Coherent Beam Combination at Interfaces via Surface Polariton Effects

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We examine theoretically the structure and properties of index gratings created by the interference pattern formed by a laser beam and a surface polariton at an interface coated with a thin layer of an optical Kerr material. Theory suggests that the energy-transfer characteristics of these gratings differ considerably from those created in the bulk material. This statement is underscored by a the prediction that energy transfer will occur for degenerate beams in the presence of a nonlinearly responding reflecting surface even though there is no photorefractive material present.

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Recently, there has been considerable interest in coherent beam combination via two-wave mixing^{1,2} utilizing laser-induced gratings³ in nonlinear optical media. In two-wave mixing, coherent energy transfer is accomplished in the following manner: A nonlinear medium is irradiated by two laser beams which form an interference pattern. This pattern, in turn, induces an index grating in the medium through its optical nonlinearity. If there is a phase lag between the induced index grating and the laser interference pattern, energy will be coherently scattered from one beam to the other. In this Letter, we examine the nature and two-wave mixing characteristics of laser-induced index gratings created at a vacuum-metal interface which is coated with a thin layer of an optical Kerr material. The properties of these index gratings⁴ at the reflecting surface (viz. metal surface) are found to differ considerably from other systems presently under study.^{1,2,5}

To date, two different approaches to two-wave mixing have been considered. Specifically, the first approach involves static gratings induced by two degenerate lasers irradiating a photorefractive crystal such as $Bi_{12}SiO_{20}$ (BSO).⁵ In this class of materials, it is found that the induced index grating lags the laser interference pattern by a phase angle nearly equal to $\pi/2$. Numerous experiments^{5,6} have demonstrated very high efficiencies for energy transfer at low laser powers in these materials. The second approach involves irradiating an optical Kerr media with two nondegenerate laser beams.¹ For this approach, the finite response time of the Kerr material, τ , ensures that any laser-induced grating will lag the interference pattern formed by the laser beams themselves. Coherent energy transfer is optimized when $\Omega \tau$ is on the order of unity, with Ω equal to the beat frequency between the two lasers. Furthermore, energy is always transferred from the high-frequency beam to the low-frequency one with the energy transfer approaching continuously to zero as $\Omega \rightarrow 0$.

Here we examine beam combination at a metal surface, where theory asserts that the existence of a welldefined resonant excitation, namely, surface polaritons, will alter the nature and enhance the wave-mixing characteristics of index gratings generated at the interface.

The system of interest consists of an interface formed by a metal and vacuum that is coated with an ultrathin layer of an optical Kerr medium whose instantaneous third-order susceptibility components are denoted by $\chi^{(3)}_{\mu\gamma\sigma\lambda}$ and whose response is accounted for by a characteristic time τ .⁷ This system is driven by two electromagnetic waves $\mathbf{E}_1(\mathbf{r},t)$ and $\mathbf{E}_2(\mathbf{r},t)$. The first of these is a laser beam whose wave vector (frequency) is $\mathbf{k}_1(\omega_1)$ which impinges on the interface at an angle θ_1 with respect to the outward normal. The second wave is a surface polariton whose wave vector (frequency) is $\mathbf{k}_2(\omega_2)$ which is propagating parallel to the surface. The beat frequency is $\Omega = \omega_1 - \omega_2$. Next, we note that if the coating thickness l satisfied $k_j l \ll 1 (j = 1, 2)$, then the electromagnetic polarization, $P_c(\mathbf{r},t)$, can be regarded as confined to the interface, i.e.,

$$\mathbf{P}_{c}(\mathbf{r},t) = l \left\{ \left| \frac{\epsilon_{c}-1}{4\pi} \right| \mathbf{E}(\mathbf{r},t) + \mathbf{P}_{\mathrm{NL}}(\mathbf{r},t) \right\} \delta(z=0^{+}).$$
(1)

In Eq. (1), ϵ_c is the linear dielectric constant of the coating and if the dynamics of the material are dominated by Debye relaxation,⁷ then the nonlinear polarization responsible for beam combination is, with $\mathbf{E}(\mathbf{r},t)$ the total electric field,

$$P_{\mathrm{NL};\mu}(\mathbf{r},t) = \frac{\chi_{\mu\gamma\sigma\lambda}^{(3)}}{\tau} \int_{-\infty}^{t} \int_{-\infty}^{t_1} dt_1 dt_2 \exp\left(\frac{t_1 - t_2}{\tau}\right) \langle E_{\gamma}(\mathbf{r},t_2) E_{\sigma}(\mathbf{r},t_2) \rangle E_{\lambda}(\mathbf{r},t_1), \qquad (2)$$

where the angle brackets represent a time average which is long compared to an optical period, but short compared to the response time of the optical Kerr material τ .

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The total field obeys the following wave equation:

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r},t) + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{E}(\mathbf{r},t) + \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{P}_{\mathrm{L}}(\mathbf{r},t) = -\frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{P}_{\mathrm{c}}(\mathbf{r},t), \qquad (3a)$$

where $\mathbf{P}_{\mathrm{L}}(\mathbf{r},t)$ is given by

$$\mathbf{P}_{\mathrm{L}}(\mathbf{r},t) = \begin{cases} 0, & z > l, \\ \int_{-\infty}^{t} dt_1 \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{\epsilon(\omega) - 1}{4\pi} \mathbf{E}(\mathbf{r},t_1) e^{-i\omega(t-t_1)}, & z < 0, \end{cases}$$
(3b)

with $\epsilon(\omega)$ the dielectric function of the metal.

To solve Eqs. (3), we introduce a Green's tensor⁸ $G_{\mu\nu}(\mathbf{r},\mathbf{r}';t-t_1)$ which satisfies the equation

$$\nabla \times \nabla \times \mathbf{G}(\mathbf{r},\mathbf{r}';t-t_1) - \frac{1}{c^2} \int dt_2 \int \frac{d\omega}{2\pi} \omega^2 \epsilon(\omega \mid z) \mathbf{G}(\mathbf{r},\mathbf{r}';t_2-t_1) e^{-i\omega(t-t_2)} = -4\pi \mathbf{I}\delta(\mathbf{r}-\mathbf{r}')\delta(t-t_1), \tag{4}$$

where $\epsilon(\omega | z) = \epsilon(\omega)\theta(-z) + \theta(z)$. Accordingly, the total electric field is given by the following:

$$\mathbf{E}(\mathbf{r},t) = \mathbf{E}_0(\mathbf{r},t) + \frac{1}{c^2} \int d^3 r_1 \int dt_1 G(\mathbf{r},\mathbf{r}';t-t_1) \frac{\partial^2}{\partial t_1^2} \mathbf{P}_c(\mathbf{r}',t_1),$$
(5)

where $E_0(\mathbf{r},t)$ is the total electric field in the absence of a nonlinear coating on the interface. To simplify matters, we shall assume that the system is infinite in the y direction and that the configuration is such that there is no y dependence in the problem. Accordingly, the fields in steady state at a point z from the interface are given by

$$\mathbf{E}(k_j\omega_j|z) = \mathbf{E}_0(k_j\omega_j|z) - l\mathbf{g}(k_j\omega_j|z0^+) \cdot \mathbf{P}_c(k_j\omega_j|0^+),$$
(6)

where $\mathbf{P}_c(k_j\omega_j|0^+)$ is the coating polarization, whose wave number (frequency) is $k_j(\omega_j)$, evaluated just above the metal $(z=0^+)$ and the reduced Green's tensor $\mathbf{g}(k_j\omega_j|z0^+)$ is given in Ref. 8.

Finally, for an s-polarized laser beam and a ppolarized surface polariton we have, with $k_1 = (\omega_1/c)\cos\theta_1$ and z > 0,

$$\mathbf{E}_{0}(k_{1}\omega_{1}|z) = E_{1}^{s}(e^{-ik}1^{z} + r_{s}(\omega_{1})e^{ik}1^{z})\mathbf{\hat{e}}_{y}, \qquad (7a)$$

$$\mathbf{E}_{0}(k_{2}\omega_{2}|z) = \left(\hat{\mathbf{e}}_{x} + i\frac{k_{2}}{\alpha_{2}}\hat{\mathbf{e}}_{z}\right)E_{2}^{p}e^{-\alpha_{2}z},$$
(7b)

where $r_s(\omega_1) = (ik_1 + \beta_1)/(ik_1 - \beta_1)$ is the Fresnel coefficient and $E_1^s(E_2^P)$ is the laser (surface polariton) electric field amplitude and $\alpha_2(\beta_2)$ is the inverse penetration length of the surface polariton into the vacuum (metal). We can obtain analytic solutions to Eqs. (6) in the limits where one wave is much more intense than the

limits where one wave is much more intense than the other. Thus, for the case in which the laser beam is intense and the surface polariton weak, we have within the nondepleted pump (laser) approximation

$$E_{x}(k_{2}\omega_{2}|z) = \frac{E_{2}^{p}}{1 + I^{ps}(\omega_{2},\omega_{1})}e^{-\alpha_{2}z},$$
 (8a)

$$E_z(k_2\omega_2|z) = \frac{ik_2}{\alpha_2} E_x(k_2\omega_2|z),$$
 (8b)

$$I^{ps}(\omega_{2},\omega_{1}) = \frac{1}{3}\xi(\omega_{2},\omega_{1}) |E_{1}^{s}|^{2} |1 + r_{s}(\omega_{1})|^{2} \frac{\alpha_{2}\beta_{2} - \epsilon(\omega_{2})k_{2}^{2}}{\epsilon(\omega_{2})\alpha_{2} + \beta_{2}} \frac{c}{\omega_{2}},$$
(9a)

$$\xi(\omega_2,\omega_1) = g\pi(\omega_2/c)l\{1 + [1 - i(\omega_1 + \omega_2)\tau]^{-1} + (1 + i\Omega\tau)^{-1}\}.$$
(9b)

Alternatively, if the laser beam is weak and the surface polariton is strong, then if we neglect depletion of the surface wave, the laser intensity is

$$E_{y}(k_{1}\omega_{1}|z) = E_{1}^{s}(e^{-ik_{1}z} + R_{s}(\omega_{1},\omega_{2})e^{ik_{1}z}),$$
(10)

where $R_s(\omega_1, \omega_2)$ is the reflection coefficient⁴ at the vacuum-metal interface coated with a thin-layer Kerr material, which is modified by the presence of the strong surface polariton, and is given by

$$R_{s}(\omega_{1},\omega_{2}) = \frac{r_{s}(\omega_{1}) - I^{sp}(\omega_{1},\omega_{2})}{1 + I^{sp}(\omega_{1},\omega_{2})},$$
(11)

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FIG. 1. The change of the x component of the electric field amplitude of a weak p-polarized surface polariton coupled to a strong s-polarized laser beam as a function of the coupling parameter.

where

 $I^{sp}(\omega_1,\omega_2)$

$$=\xi(\omega_{1},\omega_{2})|E_{2}^{p}|^{2}\left(1+\frac{1}{3}\left|\frac{k_{2}}{\alpha_{2}}\right|^{2}\right)\frac{\omega_{1}}{c}\frac{1}{ik_{1}-\beta_{1}},$$
(12a)
$$\xi(\omega_{1},\omega_{2})=g\pi\frac{\omega_{1}}{c}l\left[1+\frac{1}{1-i(\omega_{1}+\omega_{2})\tau}+\frac{1}{1-i\Omega\tau}\right].$$
(12b)

In Eqs. (9) and (12) g is the instantaneous nonlinear susceptibility, $\chi_{1122}^{(3)}$, where the Kerr material is assumed to be isotropic and possess Kleinman symmetry.⁷ An examination of these equations reveals that energy transfer will occur if $1 + I^{ps}(1 + I^{sp})$ is nearly zero for a strong bulk (surface) wave.

The salient numerical results are summarized in Figs. 1-3. Figure 1 depicts depletion of the surface polariton as a function of the effective coupling constant $\lambda = (4\pi^2/3)g |E_1^s|^2 l/\lambda_2$ at the vacuum-silver interface, in which the laser beam is strong and the Kerr medium responds instantaneously. The surface polariton can be, for example, generated by the end-fire coupling.⁹ The dielectric constant of silver is given by $\epsilon(\omega) = \epsilon_b [1 - \omega_p^2/(\omega^2 + i\omega v)]$, where $v(\omega_p)$ is the collision (plasma) frequency of the silver substrate. The attenuation of surface polariton is accounted for by v. For this case, a phase lag between the electromagnetic and index gratings is solely achieved via the response of the reflecting surface.⁴ That is, the phase lag between these two grat-



FIG. 2. Percent change in the magnitude of the x component of the electric field of a weak p-polarized surface polariton coupled to a strong s-polarized laser beam as a function of $\Omega \tau$.



FIG. 3. Amplification of a weak s-polarized laser beam by an intense p-polarized surface polariton as a function of $\Omega \tau$ for select values of the coupling parameter.

ings is set by the surface polariton response function, viz., $\mathbf{g}(k_2\omega_2|z0^+)$, and not by $\Omega\tau$, as we have assumed an instantaneous Debye relaxation to illustrate our point. An examination of this figure reveals that energy transfer to the laser beam is achieved regardless of the sign of Ω , since $\tau = 0$. More precisely, an index grating which is impressed on the ultrathin nonlinear coating by the interference pattern created by the weak surface polariton and the strong laser beam via the optical Kerr effect will always diffract the energy of a surface wave away. Another interesting feature of this figure is the dependence of the energy extraction on the sign of g. Note that energy extraction from the surface wave is significant for a coupling constant $\lambda \simeq 0.1$. For a Kerr material with $g \simeq 10^{-7}$ esu, this value can be achieved with laser-beam intensities on the order of 10 GW/cm^2 for coating thickness on the order of 1% of the wavelength of the surface polariton.

The relaxation effects are illustrated in Fig. 2, which depicts the percentage of gain, or depletion for the curves labeled (x-1), of the x-component electric field amplitude of the surface polariton versus $\Omega \tau$ for different angles of incidence of the s-polarized laser beam on a Ag surface with $\lambda = \frac{1}{3} 4\pi^2 g |E_1^s|^2 l/\lambda_2 = 10^{-2}$. Note that the direction of energy flow can switch signs as $\Omega \tau$ increases in value.

Figure 3 depicts the amplification of a weak laser beam by an intense surface polariton which resides on silver substrate as a function of $\Omega \tau$ for different values of the coupling constant, where $\lambda = 4\pi^2 g |E_1^p|^2 l/\lambda_1$. Essentially, the phase and modulus of modified reflection coefficient $R_s(\omega_1, \omega_2)$ determine the energy transfer between the surface polariton and the weak laser beam. For the case in Fig. 3, Ω is chosen to be 1 cm⁻¹ so that we are considering Kerr media with response times on the order of 30 psec.

In conclusion, theory predicts that an index grating created within the optical Kerr coating formed by a laser beam and a surface polariton of the same frequency will modify the reflectivity of the laser beam and coherently transfer energy from the surface wave to the laser beam for an instantaneous Kerr coating even though there is no photorefractive effect in the system. Furthemore, if the frequencies of the bulk and surface wave are different, then the direction of energy flow is not set just by the sign of Ω . Specifically, the phase of the effective coupling function ξ as well as the phase of the surface polariton or laser beam response function are important quantities which determine the direction of energy flow. In addition, the direction of energy flow can reverse itself as Ω increases in magnitude (with no change in sign). These features of bulk-surface wave index gratings reflect the convoluted phase structure of the response function and the finite response time of the Kerr coating. Finally, we find that the intensity of a weak bulk wave can be amplified by factors of 10^4 or more by extracting energy from a strong surface polariton, which modifies the phase and modulus of the reflection coefficient⁴ of the weak bulk wave.

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