Phase conjugation by four-wave mixing of electromagnetic surface waves

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We have studied phase conjugation by four-wave mixing of electromagnetic surface waves, both experimentally and theoretically. In the experiment, an electromagnetic surface wave was excited at the edge of a silver film by means of frustrated total internal reflection. We obtained a reflection coefficient of 10% in the conjugate wave, with an energy density in the pump waves of 5 mJ/cm^2; the fraction of the wavefront conjugated was 75%. We have investigated the mechanism whereby the permittivity grating is recorded.

1. INTRODUCTION

As shown by an example of second-harmonic generation,[1–3] involvement of electromagnetic surface waves (ESW) in nonlinear optical interactions makes it possible to raise their efficiency significantly. The production of a volume signal wave can then arbitrarily be divided into three steps. In the first, frustrated total internal reflection (FTIR)[1,4] of the original volume radiation is used to excite ESW of the same frequency at the boundary between a metal film and a dielectric. Next, because of the nonlinear interaction of the electromagnetic wave with the near-surface layers of the metal[2] or dielectric,[3] a signal-bearing ESW is formed. Finally, the latter excited a volume signal wave, which is then detected as part of the experiment.

It has recently been shown experimentally[5,6] that in a similar approach, when the middle step employs degenerate four-wave mixing (FWM) of electromagnetic surface waves, it is possible to obtain a volume wave which is counterpropagating relative to the volume signal wave. The feasibility of phase conjugation[7] by degenerate FWM of surface waves has thereby been demonstrated. As in the case of four-wave mixing of volume waves, which has of late been studied in detail,[7,8] three ESW were excited in the experiments described in Refs. 5 and 6, with two of them (the pumps) counterpropagating. Because of the nonlinear response of the metal[5] or the dielectric,[5,6] the signal and one of the pump waves records a permittivity grating. Taking it into account, the other pump wave forms a fourth ESW with a wavefront which is conjugate to the signal ESW. This conjugate wave constitutes the source of the volume wave propagating in the direction opposite that of the signal volume wave. It has been shown that a thermal mechanism is responsible for the observed effect, recording the permittivity grating. However, the FWM efficiency obtained in Refs. 5 and 6 was fairly low, and the quality of the conjugate wave was not checked. At the same time, as we shall show in the present paper, the approach described enables one to obtain a high reflection coefficient into the conjugate wave, with good quality, at relatively low pump energies. We also examine in detail the mechanism whereby the permittivity grating is recorded.

2. THEORY

Consider a metal film of thickness l (medium 2 in Fig. 1a) situated between two dielectrics. We shall neglect the imaginary part of the permitivities ε_1, ε_3 of the dielectrics, and assume that $\varepsilon_3 < \varepsilon_1 < -\operatorname{Re}\varepsilon_2$. Three p-polarized waves of frequency ω are incident at an angle θ on the upper boundary of the film $z = l$ (see Fig. 1a), with tangential field components

$$E_j = \mathscr{E}_j \exp[-i\omega t + i\mathbf{k}_j\boldsymbol{\rho} - ik_\perp(z-l)],$$
$$j = 1,\ 2,\ 3,$$
$$|\mathbf{k}_j| = k = (\omega/c)\,\varepsilon_1^{1/2}\sin\theta,$$
$$k_\perp = (\omega/c)\,\varepsilon_1^{1/2}\cos\theta,$$

where $\boldsymbol{\rho}$ is a two-dimensional coordinate in the plane of the surface E_1 and E_2 are the reference waves and are reflected in opposite directions by the surface ($\mathbf{k}_1 = -\mathbf{k}_2$; see Fig. 1b), while E_3 is the signal undergoing conjugation. Waves of the form

$$r\mathscr{E}_j \exp[-i\omega t + i\mathbf{k}_j\boldsymbol{\rho} + ik_\perp(z-l)],$$

are reflected from the surface, where r is the complex amplitude of the reflection coefficient. At angles of incidence θ greater than the total internal reflection angle for the interface between the upper and lower dielectrics, $\theta_{\mathrm{TIR}} = \arcsin(\varepsilon_3/\varepsilon_1)^{1/2}$, the tangential field component distribution in the lower medium may be written in the form

$$\tau\mathscr{E}_j \exp(-i\omega t + i\mathbf{k}_j\boldsymbol{\rho} + Qz), \qquad Q = (k^2 - \varepsilon_3\omega^2/c^2)^{1/2}.$$

We may find the values of r and τ by solving the Maxwell equations in all media, taking the boundary conditions at $z = l$ and $z = 0$ into account.[9] If the thickness l of the

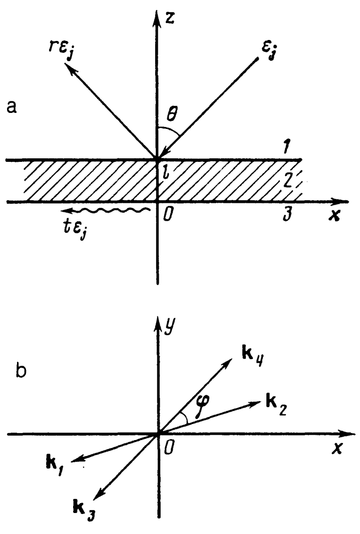

FIG. 1. Layout for excitation of electromagnetic surface waves.

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metal layer is chosen optimally, then an ESW will be excited efficiently at the lower boundary of the film for certain angles of incidence θ, as manifested by a reduction in the (intensity) reflection coefficient $R = |r|^2$ for optical beams incident on the surface.[1,4] The intensity of the electromagnetic field at the lower boundary of the metal layer then rises substantially, so that the factor

$$T = \frac{1+k^2/Q^2}{1+k^2/k_\perp^2} |\tau|^2,$$

which characterizes the increase, can reach values of the order of 10^2.

Because of dissipation of the energy of the interfering beams in the metal layer from the onset of the laser pulse, a temperature grating arises which with time is also transferred to the dielectrics. If the permittivity of the metal and the lower dielectric depend on temperature, a permittivity grating will be recorded in both media. Since the dielectric may be a liquid, with a high degree of thermal nonlinearity, the permittivity grating recorded in the lower medium elicits the greatest interest. In what follows, we devote most attention to that medium.

For the nanosecond laser pulses used in the experiment, gratings with a high spatial frequency were washed out on account of the high thermal conductivity of the metal. We therefore assume the angle φ in Fig. 1b to be small, and consider only the grating formed by interference of the signal with the passing reference wave E_1. The increment in permittivity of the lower medium, which is directly responsible for the growth of the conjugate wave, may then be written as

$$\Delta\varepsilon_3=\delta(z,\ t)\exp[i(\mathbf{k}_1-\mathbf{k}_3)\boldsymbol{\rho}].$$

When the second wave E_2 is scattered from this grating, a conjugate wave of the form

$$E_4=\mathscr{E}_4\exp[-i\omega t+i\mathbf{k}_4\boldsymbol{\rho}+ik_\perp(z-l)], \qquad \mathbf{k}_4=-\mathbf{k}_3$$

arises in the upper medium. If we replace the amplitude of the permittivity grating $\delta(z,t)$ with some effective value $\delta_{\text{eff}}(t)$ which is independent of z, then the amplitude $\mathscr{E}_4$ of the conjugate wave is easily found, using results on phase conjugation by a surface:[10,11]

$$\mathscr{E}_4=(\partial r/\partial\varepsilon_3)\delta_{\text{eff}}\mathscr{E}_2. \tag{1}$$

Differentiating the well known expression for the amplitude reflection coefficient r (Ref. 9), we can transform (1) to the form

$$\mathscr{E}_4=\frac{1}{4}\frac{ik_\perp}{\varepsilon_1 Q}\left(1+\frac{k^2}{Q^2}\right)\tau^2\delta_{\text{eff}}\mathscr{E}_2. \tag{2}$$

Given an arbitrary z-distribution for the amplitude of the permittivity grating $\delta(z,t)$, a rigorous solution of the electrodynamic problem makes it possible to find its effective value:

$$\delta_{\text{eff}}(t)=2Q\int_{-\infty}^{0}\delta(z,t)e^{2Qz}\,dz. \tag{3}$$

Note that the approximation of plane interacting waves is only valid when the transverse dimensions of the beams incident on the film are much greater than the propagation distance of the ESW along the lower boundary of the film. This condition was satisfied in our experiments.

In general, for an exact determination of the effective amplitude $\delta_{\text{eff}}(t)$, it is necessary to know the temperature profile in the lower medium during the entire laser pulse. Numerical estimates show, however, that if the pulse is several tenths of a nanosecond long, the depth of localization of the electromagnetic field in the lower medium is much greater than the thickness d of the heated layer: $1/Q \gg d$. In that event, the problem is greatly simplified, since the effective amplitude δ_{eff} is determined by the quantity of heat taking part in the writing of the grating. Transforming (3), we find

$$\delta_{\text{eff}}(t)=2Q\frac{1}{\rho c_p}\frac{\partial\varepsilon_3}{\partial T}\xi(1-R)\cos\theta[W_1(t)W_3(t)]^{1/2}, \tag{4}$$

where ρ and c_p are the density and heat capacity of the lower medium, respectively, $\partial\varepsilon_3/\partial T$ is the temperature derivative of the permittivity,

$$W_j(t)=\int_0^t I_j(t')\,dt'$$

is the instantaneous value of the energy density in the beam, $I_j = |\mathscr{E}_j|^2$ is the beam intensity, and ξ is the fraction of the energy propagating in the lower medium which is absorbed by the film. In determining ξ, it is necessary to take into account the fact that since the angle between the signal beam and reference beam E_1 is small, heat exchange between the maxima and minima of the temperature grating can be neglected. A metal film with a thickness of several hundred angstroms is heated up in a time 10^{-12}–10^{-11} sec, which means that the temperature can be taken to be constant throughout the thickness. The overall amount of thermal energy propagating in the upper and lower dielectrics is much greater than that portion remaining in the film. Making use of the foregoing, we can determine ξ, which is constant during the entire pulse:

$$\xi=[1+(\lambda_1/\lambda_3)(\chi_3/\chi_1)^{1/2}]^{-1}. \tag{5}$$

Here λ_j and χ_j are the thermal conductivity and thermal diffusivity of the dielectrics, respectively. Finding the effective amplitude δ_{eff} in this way and substituting into (2), we obtain after some manipulation an expression for the time-dependence of the conjugate beam intensity:

$$I_4(t)=\frac{1}{4}I_2(t)\frac{1}{\varepsilon_1}\left(\frac{\omega}{c}\right)^2\frac{1}{(\rho c_p)^2}\left(\frac{\partial\varepsilon_3}{\partial T}\right)^2 \times(1-R)^2T^2\xi^2W_1(t)W_3(t). \tag{6}$$

Knowing the shape of the laser pulse, we can also determine the total energy W_4 in the conjugate beam. Thus, for a pulse with a parabolic time envelope,

$$W_4=\frac{1}{12}\frac{1}{(\rho c_p)^2}\left(\frac{\partial\varepsilon_3}{\partial T}\right)^2\frac{1}{\varepsilon_1}\left(\frac{\omega}{c}\right)^2(1-R)^2T^2\xi^2W_1W_2W_3, \tag{7}$$

where W_j is the total energy density in the beam. For a Gaussian pulse, Eq. (7) remains valid, up to a factor of order unity.

3. DETERMINATION OF THE PERMITTIVITY OF SILVER AND THE THICKNESS OF THE FILM

In our experiment, electromagnetic surface waves were generated at the boundary of a thin silver film by radiation from a single-mode ruby laser with a wavelength $\lambda = 694$ nm. Metal films were prepared by vacuum deposition of silver on the diagonal face of a right-angle glass prism. A cell was attached to the prism, and when necessary it could be filled with ethanol, which was then in contact with the silver, making it possible to excite ESW not only at the silver-air interface, but at the interface between the silver and the ethanol as well. Radiation polarized in the plane of incidence passed through a side face of the prism and fell on the metal film from the glass side. The angle of incidence θ of the beam on the film was varied during the experiment.

It can be seen from the previous section that the efficiency of FWM depends strongly on T, specifically on the permittivity of all three media and the thickness of the film l. For a quantitative comparison of experimental and theoretical results, it is therefore desirable to determine these quantities to fairly high accuracy. Since the permittivity ε_2 of the metal, especially its imaginary part, depends strongly on the deposition conditions, the data from various papers show a large spread and cannot be used. Direct methods for determining the thickness of the metal film also fail to provide the requisite accuracy.

To determine these quantities, we have measured the transmission coefficient of the metal film when it is illuminated. Furthermore, we have measured the angular dependence of the reflection coefficient R of p-polarized radiation from the metal film with FTIR geometry at angles of incidence corresponding to efficient excitation of ESW at the silver-ethanol interface. For the film used in subsequent experiments, this dependence is shown by the points plotted in Fig. 2. Using the value of the angle at which the reflection coefficient has a minimum, one can determine Re ε_2. The thickness of the film is determined using its experimentally measured transmission coefficient. Finally, by varying the magnitude of Im ε_2, it is possible to match the calculated angular dependence of R (solid curve in Fig. 2) to the experimental data. As a result, we obtained the values $\varepsilon_2 = -19.8 + 0.9i$ and $l = 475$ Å. Note that the refractive index of the glass and the reference are 1.57 and 1.36 respectively, and the thickness of the film as determined by weighing lay in the range 400–500 Å.

In Fig. 2, besides the reflection coefficient, we have also plotted the angular dependence of the quantity T (dashed curve). The maximum value of T is seen to be equal to 70. Note that when an ESW is excited at the silver-air interface, as shown by the calculations, the same film can give $T = 225$.

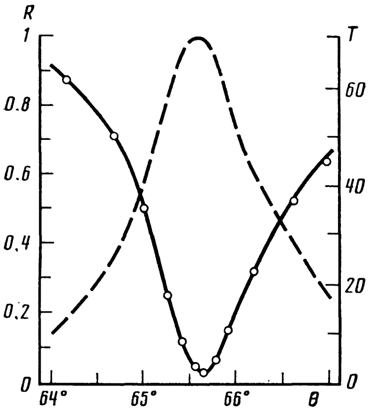

FIG. 2. Angular dependence of the factor T and the reflection coefficient R.

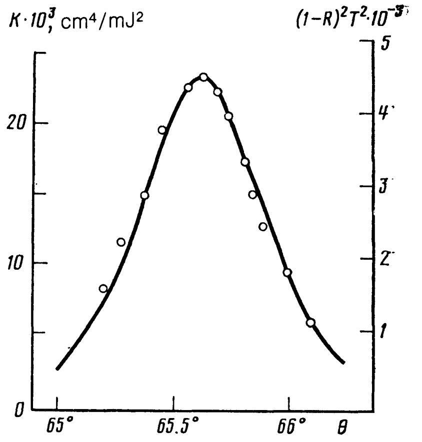

FIG. 3. Angular dependence of efficiency of four-wave mixing.

4. ANGULAR DEPENDENCE OF FOUR-WAVE MIXING EFFICIENCY. TEMPORAL BEHAVIOR OF THE CONJUGATE BEAM

For our study of the angular dependence of FWM efficiency, three p-polarized beams were incident on the film. In each laser burst, all beams had the same angle of incidence θ on the film, with the E_1 and E_2 reference beams reflected from the surface in counterpropagating directions ($\mathbf{k}_1 = -\mathbf{k}_2$). The angle between E_1 and the signal beam was fixed at 1.5° (in glass). The ratio of beam energy densities was held constant in this experiment, with $W_3{:}W_1{:}W_2 = 1{:}10{:}13$. We measured the energy density of the conjugate beam propagating in the direction opposite that of the signal beam. The points in Fig. 3 are experimentally measured values of FWM efficiency $K = W_4/W_1W_2W_3$ with ESW excited at the silver-ethanol interface. The measurements were carried out with low energy densities in the reference beams, since at high W, the cubic dependence of signal energy on laser beam energy breaks down; we shall subsequently have more to say about this.

In the second section of this paper, it was suggested that the permittivity grating in the ethanol was mainly responsible for generating the conjugate wave, and that the grating formed in the metal could be neglected. To test this assertion experimentally, we measured the angular dependence of FWM efficiency with the ethanol missing, exciting an ESW at the silver-air interface. The conjugate wave is then produced by the permittivity grating formed in the metal. The maximum efficiency was 15 times lower than with the ethanol present. At the same time, calculations show that if the effect were due to the grating in the metal for the silver-ethanol interface, then FWM efficiency would, on the contrary, be an order of magnitude higher. Thus, we conclude that with the ethanol in place, the contribution of the grating in the metal can be neglected for nanosecond pulses. According to (7), FWM efficiency should then be proportional to $(1-R)^2T^2$, which has been calculated and plotted in Fig. 3 as the solid curve. Clearly, the theory provides a good description of the angular dependence of FWM efficiency.

Equation (7) also enables us to compare the maximum efficiency with theoretical predictions. Unfortunately, we did not know the thermophysical constants of the glass, which are required for an accurate calculation of the fraction ξ of the energy which takes part in writing the permittivity

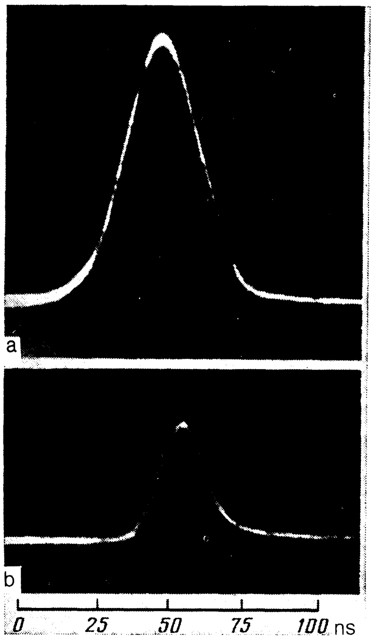

FIG. 4. Pulse shapes for incident (a) and conjugate (b) beams.

grating. These constants vary only slightly with the type of glass used, and we can estimate ξ to within a factor of two as $\xi = 0.2$–0.4. The same quantity can be calculated using the experimetal data. The value thus obtained, $\xi = 0.24$, lies within the expected range, i.e., there is reasonable quantitative agreement between theory and experiment.

Also of interest is the temporal history of the conjugate wave pulse. Figure 4 shows pulse shapes for the incident (Fig. 4a) and conjugate (Fig. 4b) beams. The incident pulse duration at the half-power points was 27 nsec. The conjugate pulse was shortened to 17 nsec, and had a noticeable time delay relative to the incident pulse (see Fig. 4). Simultaneous recording of both pulses makes it possible to calculate both the instantaneous energy density $W(t)$ in the incident beam (solid curve in Fig. 5) and the instantaneous reflection coefficient in the conjugate wave $\eta(t) = I_4(t)/I_3(t)$ (dotted curve in Fig. 5). Since according to (6), the reflection coefficient is proportional to the square of the instantaneous energy density, the dashed curve in Fig. 5 has also been plotted to show the time dependence of $W^2(t)$. It can be seen that the observed temporal behavior of the reflection coefficient $\eta(t)$ is in fair agreement with the theoretical predictions.

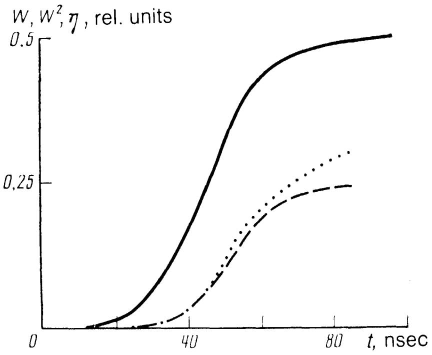

FIG. 5. Time dependence of laser beam eneregy $W(t)$ (solid curve), its square $W^2(t)$ (dashed curve), and instantaneous coefficient of reflection into a conjugate wave $\eta(t)$ (dotted curve).

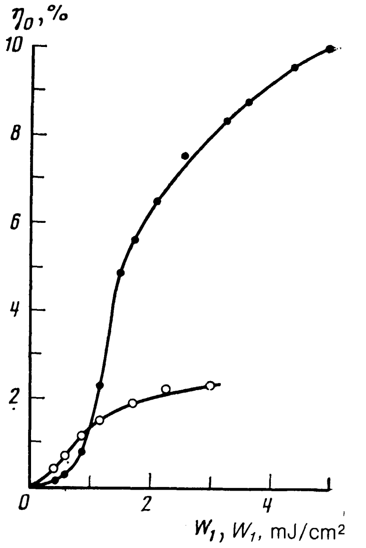

FIG. 6. Energy dependence of the conjugate-wave reflection coefficient η_0: filled circles correspond to $\theta = 65.2°$, open circles to $\theta = 65.6°$.

5. ENERGY DEPENDENCE OF THE REFLECTION COEFFICIENT IN THE CONJUGATE WAVE

One of the most important characteristics of the phase conjugation phenomenon is the absolute value of the energy reflection coefficient in the conjugate wave, $\eta_0 = W_4/W_3$. In Fig. 6, we show how this quantity depends on the energy density W_1 of the first pump wave at angles of incidence $\theta = 65.2°$ to $65.6°$, the latter angle corresponding to the highest efficiency of ESW excitation (see Figs. 2 and 3). For small W_1, according to (7), the reflection coefficient η_0 increases quadratically with W_1. This behavior no longer holds, however, for large W_1. The reason for this is that up to this point, in calculating the variation of the permittivity in the lower medium, we have not taken into account any transversely homogeneous decrease due to an overall temperature elevation in the lower dielectric. Consequently, at the end of the laser pulse, the values of R and T, which depend strongly on ε_3, can differ substantially from R and T at the beginning of the pulse. This can be shown graphically in the following way. As the length of the laser pulse increases, the curves in Fig. 2 are shifted to the left, with practically no change in shape; the size of this shift increases with increasing reference beam energy. Therefore, for $\theta = 65.6°$, the time-averaged FWM excitation efficiency is a maximum only at low reference-beam energy, and it decreases with increasing energy. At an angle of incidence $\theta = 65.2°$, the average ESW excitation efficiency first increases and then also decreases. Thus, when the angle of incidence of the beams on the metal film is reduced, the experiment value of η_0 increases (see Fig. 6).

6. QUALITY OF THE CONJUGATE WAVE

In studying another important characteristic of phase conjugation, the quality of the conjugate wave, we used standard methods employing a phase plate and beam splitter.[7,11] Our measurements have shown that the phase conjugation efficiency with uniform reference-wave intensity in the interaction region is fairly high, about 75%. It is important that the quality of the conjugate wave not be degraded as the reference-wave energy and the reflection coefficient η_0 are increased. Since the conjugate wave has an angular spectrum of finite width, it is important to ascertain the limiting width which will have no effect on phase conjugation quality. To do this, we have measured the power reflection coefficient η_0 as a function of the angular offset of the signal wave, for fixed angle of incidence θ of the pump waves. The most important factor is the deviation of the signal beam from its optimum orientation in the plane of incidence. This changes the angle of incidence θ of the beam at the film, which leads to a reduction in ESW excitation efficiency by the signal. As a result, the reflection coefficient η_0 is halved by an angular offset of 0.5°. Deviation of the signals beam perpendicular to the plane of incidence, if it increases the angle between the signal and first reference waves, also reduces η_0. This occurs because of the increased spatial frequency of the interference pattern and increased heat transfer between the maxima and minima of the temperature grating. Thus, when the angle between E_3 and E_1 increases from 1.5° to 7° (in glass), the value of η_0 decreases by a factor of 3.5. Since as a rule, the angular size of the signal wave is no more than 0.1°, the dependence of the reflection coefficient on the angular offset of the signal beam does not affect the quality of the conjugate wave.

7. CONCLUSION

We have shown in this work that the phase conjugation method we have described enables one to obtain sizable reflection of the conjugate wave without deterioration of quality. We have obtained a reflection coefficient $\eta_0 = 10\%$, with a low pump-beam energy density of 5 mJ/cm^2 (see Fig. 6). The thermal nonlinearity mechanism has also been extensively used for degenerate four-wave mixing of volume waves, making a comparison of the two methods possible. The expression in (6) for the efficiency of phase conjugation involving ESW differs from its counterpart for FWM of volume waves (see Sec. 7.2.3 of Ref. 7) by a factor $T^2\xi^2$. Consequently, for the same pump-beam energy densities, the reflection coefficient η_0 in our case should be more than two orders of magnitude higher, which has been verified experimentally. This leads us to believe that there are experimental situations in which the phase conjugation method described could successfully supplant degenerate four-wave mixing of volume waves.

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