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Nonlinear Excitation of Surface Polaritons

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We show that nonlinear excitation of surface-polariton waves by optical mixing should be possible. The excited surface waves can be easily detected by either the prism-coupling method or coherent scattering from wave mixing. Practical numerical examples are given.

Surface physics has recently attracted a great deal of attention. Accordingly, there has also been increasing interest in the problem of surface polaritons. The methods of investigation of surface polaritons are however limited. So far, surface polaritons and plasmons have been studied by inelastic electron diffraction,[1] by attenuated total optical reflection,[2] and by Raman scattering.[3,4] Optical second-harmonic generation has also been used to probe the coupling between the fundamental field and the surface plasmons.[5] One would expect that surface polaritons can also be nonlinearly excited by optical mixing of two laser beams. In this Letter, we show that this is indeed possible with ordinary pulsed dye lasers. We present for the first time a formulated theory of nonlinear excitation of surface polaritons. This theory can be easily extended to other problems involving nonlinear coupling of surface polaritons with bulk electromagnetic waves. We propose to detect the excited surface polaritons by either the prism method in which the surface waves are coupled out by a prism or the wave-mixing scheme in which the surface waves coherently scatter a probe beam.

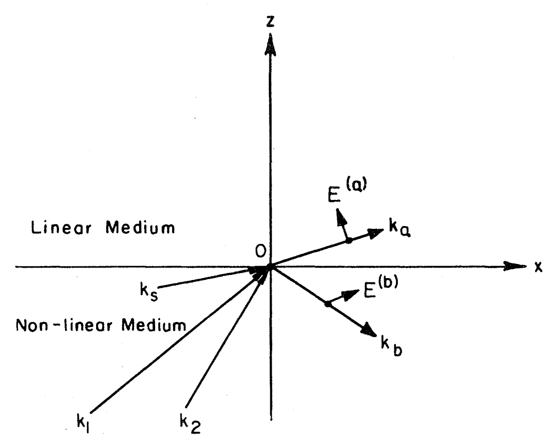

FIG. 1. Difference-frequency generation at a boundary surface. The wave vectors of incoming pump waves, the nonlinear polarization wave, and the difference-frequency waves are shown.

Our derivation is a nontrivial extension of the theoretical treatment of Bloembergen and Pershan[6] on interaction of light waves at the boundary of a nonlinear medium. Consider a plane surface at $z=0$ separating a linear medium at $z>0$ from a nonlinear medium at $z<0$. The linear dielectric constants for the linear and nonlinear media are ϵ_a and ϵ_b, respectively. Two incoming laser beams with $\vec{E}_1=\vec{\mathcal{E}}_1\exp(i\vec{k}_1\cdot\vec{r}-i\omega_1 t)$ and $\vec{E}_2=\vec{\mathcal{E}}_2\exp(i\vec{k}_2\cdot\vec{r}-i\omega_2 t)$ generate in the nonlinear medium a nonlinear polarization $\vec{P}^{(2)}(\omega=\omega_1-\omega_2)=\overleftrightarrow{\chi}^{(2)}:\vec{E}_1\vec{E}_2$, where $\overleftrightarrow{\chi}^{(2)}(\omega=\omega_1-\omega_2)$ is the nonlinear susceptibility. (The theory can be easily extended to sum-frequency generation.) The difference-frequency generation is governed by the equations

$$[\nabla\times(\nabla\times)-\omega^2\epsilon(\omega)/c^2]\vec{E}(\omega)=(4\pi\omega^2/c^2)\vec{P}^{(2)}(\omega),\quad \nabla\cdot(\epsilon\vec{E}+4\pi\vec{P}^{(2)})=0. \tag{1}$$

We are interested in a TM wave ($E_y=0$ and $P_y^{(2)}=0$) propagating in the x-z plane as shown in Fig. 1.

Consider first the case of infinite plane waves. The solution of Eq. (1) with appropriate boundary conditions is,[6] for $z>0$,

$$\vec{E}^{(a)}=\vec{\mathcal{E}}_0^{(a)}\exp(i\vec{k}_a\cdot\vec{r}-i\omega t),\quad \vec{E}^{(b)}=[\vec{\mathcal{E}}_0^{(b)}\exp(i\vec{k}_b\cdot\vec{r})+\vec{\mathcal{E}}_p\exp(i\vec{k}_s\cdot\vec{r})]\exp(-i\omega t), \tag{2}$$

where $\vec{k}_s=\vec{k}_1-\vec{k}_2$, $k_{a,b}^2=k_0^2\epsilon_{a,b}$, $k_0\equiv\omega/c$, $k_{ax}=k_{bx}\equiv k_x$ (assumed real), and

$$\vec{\mathcal{E}}_0^{(a)}=\frac{-4\pi(\hat{x}k_{az}-\hat{z}k_x)}{(\epsilon_a k_{bz}+\epsilon_b k_{az})(k_s^2-k_b^2)}[P_x^{(2)}(k_s^2-k_b^2)-(k_{bz}+k_{sz})(k_{sz}P_x^{(2)}-k_xP_z^{(2)})],$$
$$\vec{\mathcal{E}}_0^{(b)}=\frac{4\pi(\hat{x}k_{bz}+\hat{z}k_x)\epsilon_a}{\epsilon_b(\epsilon_a k_{bz}+\epsilon_b k_{az})(k_s^2-k_b^2)}\left[P_x^{(2)}(k_s^2-k_b^2)-\left(\frac{\epsilon_b}{\epsilon_a}k_{az}-k_{sz}\right)(k_{sz}P_x^{(2)}-k_xP_z^{(2)})\right], \tag{3}$$
$$\vec{\mathcal{E}}_p=-4\pi\epsilon_b^{-1}(k_s^2-k_b^2)^{-1}\{\hat{x}[-P_x^{(2)}k_{bz}^2+P_z^{(2)}k_xk_{sz}]+\hat{z}[P_x^{(2)}k_xk_{sz}+P_z^{(2)}(k_{sz}^2-k_b^2)]\}.$$

In Eq. (2), the $\vec{\mathcal{E}}_0$ terms and the $\vec{\mathcal{E}}_p$ term are, respectively, the homogeneous and the particular solutions of Eq. (1). The dielectric constants, ϵ's, are in general complex.

For illustration, let us assume the ϵ's real for the moment. Then, if k_x is larger than k_a and k_b, both k_{az} and k_{bz} become purely imaginary. The homogeneous solution now corresponds to a wave propagating along $\hat{x}$ but bounded to the boundary surface at $z=0$. We then have a surface wave excited by nonlinear optical mixing. The amplitude of the excited surface wave is a divergent maximum when $\epsilon_a k_{bz}+\epsilon_b k_{az}=0$. This equality can be readily transformed into

$$k_x^2-k_0^2\epsilon_a\epsilon_b/(\epsilon_a+\epsilon_b)=0. \tag{4}$$

We recognize that Eq. (4) is just the usual expression for the dispersion of surface polaritons.[7] The criterion for the existence of surface polaritons is $k_x>k_{a,b}$, or $\epsilon_a\epsilon_b/(\epsilon_a+\epsilon_b)>\epsilon_{a,b}$. It can be satisfied only if either ϵ_a or ϵ_b is negative. If $\epsilon_b<0$, then we must have $|\epsilon_b|>\epsilon_a$. This happens in the *Reststrahlung* phonon or exciton bands of the nonlinear medium.

More generally, ϵ's and hence k's are complex. The resonant excitation of the surface wave now occurs at

$$k_x^2=k_0^2\,\mathrm{Re}[\epsilon_a\epsilon_b/(\epsilon_a+\epsilon_b)]. \tag{5}$$

The amplitude of the resonantly excited surface wave is now finite and the linewidth is proportional to $\mathrm{Im}[\epsilon_a\epsilon_b/(\epsilon_a+\epsilon_b)]$.

However, the excited surface wave discussed above is actually a driven wave which only exists in the presence of excitation. In general, there is also a free surface wave $\vec{E}_f$ coexistent with the driven wave. From Eq. (1) for given ω, we find

$$\vec{E}_f^{(a)}=\vec{\mathcal{E}}_f^{(a)}\exp(iK_xx-i\omega t-\alpha_a z),\quad \vec{E}_f^{(b)}=\vec{\mathcal{E}}_f^{(b)}\exp(iK_xx-i\omega t-\alpha_b z), \tag{6}$$

where $K_x=K_x'+iK_x''$ is a complex quantity obeying the dispersion relation for free surface polaritons, e.g., $K_x^2=k_0^2\epsilon_a\epsilon_b/(\epsilon_a+\epsilon_b)$, α_a and α_b are defined by $\alpha_a=(K_x^2-k_a^2)^{1/2}$ and $\alpha_b=-(K_x^2-k_b^2)^{1/2}$, and the amplitudes $\vec{\mathcal{E}}_f^{(a)}$ and $\vec{\mathcal{E}}_f^{(b)}$ are to be determined by boundary conditions. Unlike the driven wave, this free surface wave can propagate even in regions without external excitation. It is the thermally excited free surface polaritons which contribute to the observed spontaneous Raman scattering by surface polaritons.[3,4] In the case of coherent excitation by continuous, infinite, plane waves, however, we should have $\vec{E}_f=0$.

Now, suppose the nonlinear excitation has a finite cross section on the surface. We assume a simple case with $|P^{(2)}|=$ const for $0\leq x\leq l$ and

$|P^{(2)}|=0$ otherwise. We also neglect the free wave propagating along $-\hat{x}$ and assume k_{sx} being close to K_x' so that $k_{az}\cong i\alpha_a$ and $k_{bz}=-i\alpha_b$. Then, since the surface wave is the sum of the free wave of Eq. (6) and the driven wave of Eq. (2), we find

$$\begin{aligned}\vec{E}^{(a,b)} &= 0 \text{ for } x\leqslant 0,\\ &= \vec{\mathscr{E}}_0[\exp(i\Delta k_x x)-\exp(-K_x''x)]\exp[i(K_x'x-\omega t)-\alpha_{a,b}z] \text{ for } 0\leqslant x\leqslant l,\\ &= \vec{\mathscr{E}}_0[\exp(i\Delta k_x a)-\exp(-K_x''a)]\exp[i(K_x'x-\omega t)-K_x''(x-a)-\alpha_{a,b}z] \text{ for } x\geqslant l,\end{aligned} \tag{7}$$

where $\Delta k_x \equiv k_{sx}-K_x'$ is the phase mismatch along $\hat{x}$. For $0\leqslant x\leqslant l$, the $\exp(-K_x''x)$ term comes from the free wave, but for $x\geqslant l$, the entire field is a free wave.

The above result is in close analogy to the familiar result for sum- or difference-frequency generation in the bulk.[8] With reflection of surface waves at $x=0$ and l neglected, the generated surface wave grows with x starting from $x=0$. Since we can transform the denominator $(\epsilon_a k_{bz}+\epsilon_b k_{az})$ in $\vec{\mathscr{E}}_0$ into the form

$$[(\epsilon_a^2-\epsilon_b^2)/(\epsilon_a k_{bz}-\epsilon_b k_{az})]\,2K_x'(-\Delta k_x+iK_x''),$$

we have for $0\leqslant x\leqslant l$

$$\vec{E}\propto(-\Delta k_x+iK_x'')^{-1}[\exp(i\Delta k_x x)-\exp(-K_x''x)]\exp[i(K_x'x-\omega t)-\alpha_{a,b}z],$$

which resembles the result for sum- or difference-frequency generation in a lossy medium. If $K_x''=0$, then the amplitude of the surface wave should grow linearly with x as expected. If $K_x''x\gg 1$, then the free-wave contribution can be neglected in comparison with the driven wave. In the region of $x\geqslant l$ we have only a free surface wave propagating with an attenuation length $1/K_x''$. In general, the excitation $P^{(2)}$ is a function of both x and y, and k_{sx} is not necessarily close to K_x'. It is then more rigorous and convenient to solve the problem by Fourier decomposition of $P^{(2)}$. In a similar manner, we can find the surface wave generated by a pulse of excitation. Time-resolved detection of surface waves with short pulsed excitation enables us to measure directly the lifetimes of surface polaritons.

We note that with $\vec{k}_1$ and $\vec{k}_2$ independently adjustable, the nonlinear excitation method has much more flexibility than the linear excitation method. In general, we can have the exciting laser beams come in from either side of the surface.

Having generated the surface wave, how do we detect it? The first obvious method is to use the prism- or grating-coupling scheme.[2] Consider the case of a prism coupler sitting in air ($\epsilon_a=1$) at a distance d away from the surface of the nonlinear medium. Let ϵ_c be the dielectric constant of the prism. Then, we find the energy flux of the coupled-out radiation from the surface wave to be

$$P(\omega)=\frac{2ck_0^2\epsilon_c^{3/2}T}{\pi(\epsilon_c-1)(k_x^2-k_{az}^2\epsilon_c)}\int|E_x^{(a)}(z=d)|^2\,dx\,dy, \tag{8}$$

where k_{az} is purely imaginary, T is the transmission coefficient from the prism to the air, and $E_x^{(a)}$ is obtained, for example, from Eq. (7) for the simple case we discussed.

The second method of detection is to monitor coherent scattering of a probe beam from the excited surface wave. This falls into the general category of four-wave mixing.[9] In the present case, the surface wave $E(\omega)$ beats with the probing field $E_3(\omega_3)$ in the nonlinear medium and creates a nonlinear polarization

$$\vec{P}^{(2)}(\omega_4=\omega_3+\omega)=\overleftrightarrow{\chi}^{(2)}(\omega_4=\omega_3+\omega):\vec{E}_3(\omega_3)\vec{E}^{(b)}(\omega)$$

which then generates a new field $\vec{E}_4(\omega_4)$. From the usual parametric approximation, we find the power output of the coherent scattering signal at ω_4 to be

$$P(\omega_4)=\frac{2\pi\omega_4^4[\mathscr{E}_b'(\omega_4)T]^{1/2}}{c^3k_{4z}^2}\,|\overleftrightarrow{\chi}^{(2)}:\int_{-\infty}^0\vec{E}(\omega_3)\vec{E}^{(b)}(\omega)\exp(-i\vec{k}_4\cdot\vec{r})dz|^2\,dx\,dy, \tag{9}$$

where T is the transmission coefficient from the nonlinear medium to air (the linear medium).

We now give two numerical examples, one for surface phonon polariton and one for surface exciton polariton, to illustrate the feasibility of the proposed methods. We consider first the surface phonon polariton associated with the TO phonon at 367.2 cm^{-1} in GaP. Let the surface polariton frequency be 380 cm^{-1}. The other relevant parameters are $\epsilon_b' = -11.7$, $\epsilon_b'' = 1$, $\epsilon_a(\text{air}) = 1$, $\omega_1 = 17\,000\ cm^{-1}$, $\omega_2 = 16\,620\ cm^{-1}$, and $\chi^{(2)}(\omega = \omega_1 - \omega_2) = 1.57 \times 10^{-6}$ esu.[10] We assume a peak power of 40 kW and a cross section of $2 \times 2\ mm^2$ for each of the two laser beams at ω_1 and ω_2. In the prism method, we can use a CsI prism ($\epsilon_c = 2.9$) situated at 1 μm away from the plane surface of GaP. Assuming $T = 1$, we find from Eq. (8) with $k_{az} = 0$ that the coupled-out infrared radiation at 380 cm^{-1} will have a peak power of ~0.4 W. Using the four-wave mixing scheme and assuming a probing beam of 40-kW peak power and $2 \times 2\ mm^2$ cross section, we obtain from Eq. (9) with $k_{az} = 0$ a coherent scattering signal at ω_4 of $\sim 2 \times 10^{-7}$ W peak power.

Consider next the surface exciton polariton associated with the 3.209-eV transverse exciton in CuCl. We choose the surface-polariton frequency at 3.205 eV with $\epsilon_b' = -9.9$, $\epsilon_b'' = 0.13$, $\epsilon_a(\text{air}) = 1$, $\omega_1 = \omega_2 = 12\,625\ cm^{-1}$, and $\chi^{(2)} = 2.5 \times 10^{-6}$ esu,[11] and again assume a peak power of 40 kW and a cross section of $2 \times 2\ mm^2$ for each of the laser beams at ω_1, ω_2, and ω_3. We then find with a quartz-prism coupler a second harmonic output of ~45 W at $2\omega_1$. In the four-wave mixing scheme, the expected coherent scattering signal at ω_4 is 2×10^{-7} W.

These numerical examples show that nonlinear excitation of surface polaritons can be easily observed. Experimental verification of our proposal is presently in progress. As a final comment, we note that the formalism presented in this paper can be used as a basis for treating other problems of nonlinear interaction of surface waves and bulk waves.

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