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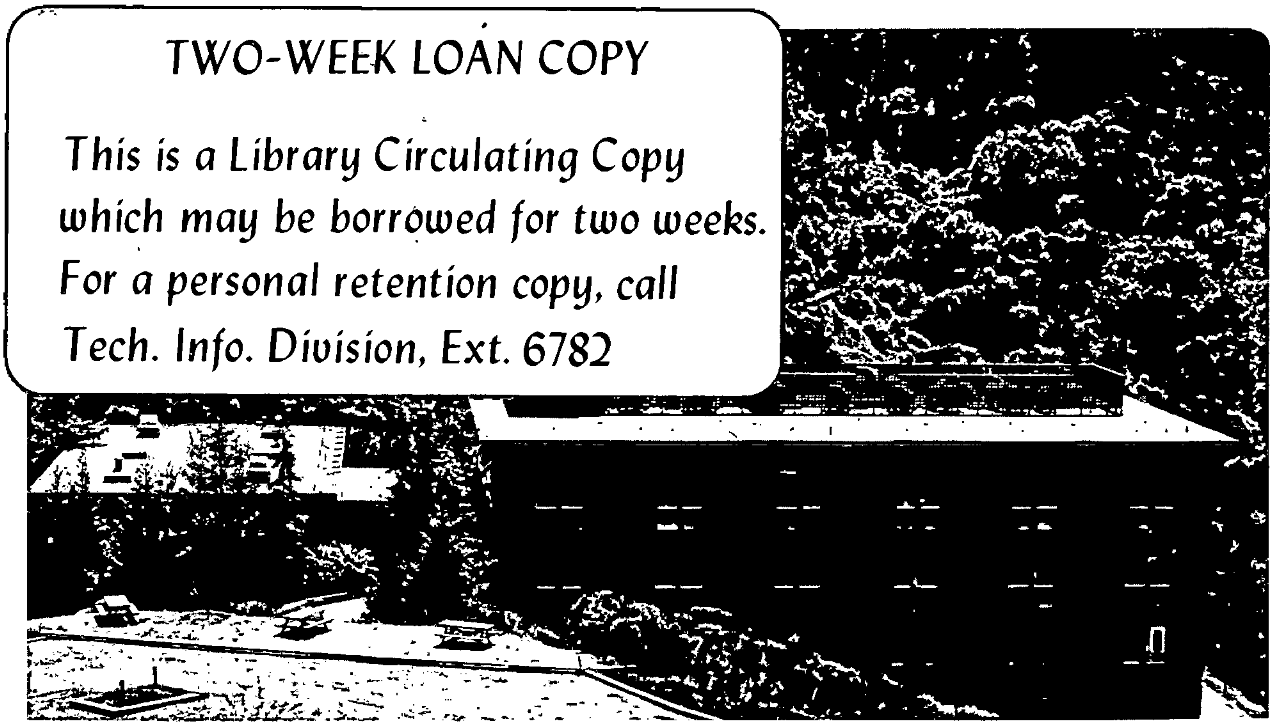

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COHERENT SECOND HARMONIC GENERATION BY COUNTER-PROPAGATING SURFACE PLASMONS

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ABSTRACT

We have observed second harmonic generation by counter-propagating surface plasmon waves. The output is in the form of a well collimated beam along the surface normal. The results are in excellent agreement with theoretical prediction.

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Nonlinear interaction of surface plasmon waves is a topic of current interest.[1-4] It is readily observable because of the high surface plasmon intensities achievable through confinement of the waves to the surface. Second harmonic generation by surface plasmons was first demonstrated by Simon et al.[2] Here, we report an experiment on coherent second harmonic generation by counter-propagating surface plasmon waves. In a bulk nonlinear medium, counter-propagating waves will lead to second harmonic generation in all directions. On the surface, however, because of the requirement of conservation of wavevector along the surface, counter-propagating surface waves generate coherent second harmonic waves only in the direction perpendicular to the surface. This peculiar effect has been pointed out earlier by several authors in a number of theoretical studies.[4] Experimentally, on the other hand, only in optical waveguides has the second harmonic generation by counter-propagating waves been studied.[5]

In our experiment, the surface plasmons along the quartz-silver interface were excited by the Kretschmann geometry[6] as shown in Fig. 1(a). It is the quartz nonlinearity which is mainly responsible for the second harmonic generation. The theoretical description follows naturally from the general treatment of second harmonic generation with boundary conditions.[1,7] Consider the case where the crystalline quartz is oriented with its $\hat{a}$-axis along $\hat{x}$ and $\hat{b}$ along $\hat{z}$. The two surface plasmon waves $\vec{E}_{s\pm} = (\mathcal{E}_{sx\pm}\hat{x} + \mathcal{E}_{sz\pm}\hat{z})\exp(\pm\, ik_x x + \beta z - i\omega t)$ generate a nonlinear polarization in quartz

$$\vec{P}^{(2)} = \hat{x}\chi_{11}^{(2)}(\mathcal{E}_{sx+}\mathcal{E}_{sx-} - \mathcal{E}_{sz+}\mathcal{E}_{sz-})\exp(2\beta z - i2\omega t) \qquad (1)$$

where $\chi_{11}^{(2)}$ is the nonlinear susceptibility of quartz. The magnitudes of

$|\mathcal{E}_{s\pm}|^2$ are related to the incoming laser beam intensities $|\mathcal{E}_{\pm}|^2$ from the two sides of the prism through the Fresnel coefficient T [8]

$$|\mathcal{E}_{s\pm}|^2 = T|\mathcal{E}_{\pm}|^2 . \tag{2}$$

Then, from the Maxwell equations with the proper boundary conditions and with $\vec{P}^{(2)}(2\omega)$ in Eq. (1) as the source term, we find that the output second harmonic field in quartz is $E(2\omega) \propto \exp[-\,ik_q(2\omega)z - i2\omega t]$ and its power is given by[1]

$$\mathscr{P}(2\omega) = \frac{c\varepsilon_q^{1/2}}{2\pi} A|F|^2 \left| \frac{4\pi(2\omega/c)^2 P_x^{(2)}}{4\beta^2 + (2\omega/c)^2 \varepsilon_q} \right|^2 . \tag{3}$$

$$F = \frac{\varepsilon_m k_q}{\alpha_m} \left\{ \frac{(\varepsilon_m k_g + i\varepsilon_g \alpha_m)(\alpha_m + 2\beta) + (\varepsilon_m k_g - i\varepsilon_g \alpha_m)(2\beta - \alpha_m) e^{-2\alpha_m d}}{(\varepsilon_m k_g + i\varepsilon_g \alpha_m)(-\varepsilon_m k_q + i\varepsilon_q \alpha_m) + (\varepsilon_m k_g - i\varepsilon_g \alpha_m)(\varepsilon_m k_q + i\varepsilon_q \alpha_m) e^{-2\alpha_m d}} \right\}$$

$$k_g = 2\omega\varepsilon_g^{1/2}/c \quad , \quad k_q = 2\omega\varepsilon_q^{1/2}/c \quad , \quad i\alpha_m = 2\omega\varepsilon_m^{1/2}/c$$

where ε_g, ε_q, and ε_m are dielectric constants of glass, quartz, and metal respectively evaluated at 2ω, A is the beam overlapping area at the interface, and d is the silver film thickness.

The experimental arrangement is shown in Fig. 1(b). A dye laser oscillator-amplifier system, with NK-199 in acetone, pumped by a Q-switched ruby laser, provided 7-mJ, 20-nsec laser pulses at 10 pulses/minute. The laser beam was tuned to 7456 Å with a linewidth less than 1 cm^{-1}, and was linearly polarized in the $\hat{x} - \hat{z}$ plane. The sample assembly was mounted on a rotary table, and consisted of an equilateral Schott SF-10 glass prism, a layer of α-bromonapthalene as index matching fluid, and a ~ 500 Å silver film evaporated on a crystalline quartz substrate. The dye laser beam was directed to the sample assembly as shown in Fig. 1(b). In order to provide a counter-propagating surface plasmon wave, the laser beam exiting from the prism was reflected back onto itself. Intensities of the two beams and the efficiency of surface plasmon excitation were monitored through beam splitters in the laser beams. The second harmonic signal was detected on the quartz side along the surface normal direction.

The experimental results are shown in Fig. 2 in comparison with theory, with the second harmonic signal plotted as a function of the angle θ. Each data point represents an average of 10 shots. The observed maximum occurs, as expected, at the angle where the counter-propagating surface plasmon waves were optimally excited, and the width of the peak is approximately the width of the surface plasmon resonance observed in the attenuated total reflection (ATR) measurement.[6] The theoretical curve in Fig. 2 was calculated from Eq. (3) using Fresnel coefficients deduced from direct fitting of the ATR surface plasmon resonance spectra. Variation of the beam cross-section A with the angle θ was also taken into account. Then, apart from a normalization constant, there is no other adjustable parameter in the theoretical calculation. Figure 2 shows a very good agreement between theory and experiment. The slight shift of the theoretical curve relative to the data points can be easily accounted for by a possible 0.2° wedge between the quartz and prism faces.

We also verified in the experiment that the generated second harmonic beam was highly directional along the surface normal with a divergence of ~ 1 mrad, and was linearly polarized along $\hat{x}$. It disappeared when one of the counter-propagating laser beams was blocked, or when the laser beam was made transverse electric so that the surface plasmons could no longer be excited, or when the quartz crystal was replaced by a glass substrate. From Eq. (3), we predict a second harmonic output $\mathscr{P}(2\omega) = 5 \times 10^{-25} \mathscr{P}_+(\omega) \mathscr{P}_-(\omega)/A$ in cgs units. In our experiment, with $A \cong 0.25\ \text{cm}^2$, $\mathscr{P}_+(\omega) = 3.5 \times 10^5$ W , and $\mathscr{P}_-(\omega) = 1.7 \times 10^5$ W , $\mathscr{P}(2\omega)$ corresponds to 2×10^4 photons/pulse. Taking into account the collection efficiency of our detection system, we estimate that our observed signal was $\sim 10^4$ photons/pulse.

The theory also predicts the generation of a second harmonic beam propagating out along the surface normal from the glass prism side. However, this beam in passing through the silver film is attenuated by a factor of $\exp(-\ 2\alpha_m d)$. Furthermore, in our experiment, a sizable background arising from second harmonic generation in the glass prism appeared to be stronger than the predicted signal. Our search for such a signal therefore has not been successful.

In conclusion, we have shown that nonlinear interaction of two counter-propagating surface plasmon waves can lead to the generation of a bulk second harmonic wave propagating out from the surface along the surface normal. In terms of language of photons, this would mean that two counter-propagating photons can annihilate each other and create a second harmonic photon propagating in the orthogonal direction. Since the effect is easily observed, it may be used to study second-order nonlinear optical properties of thin crystalline films.

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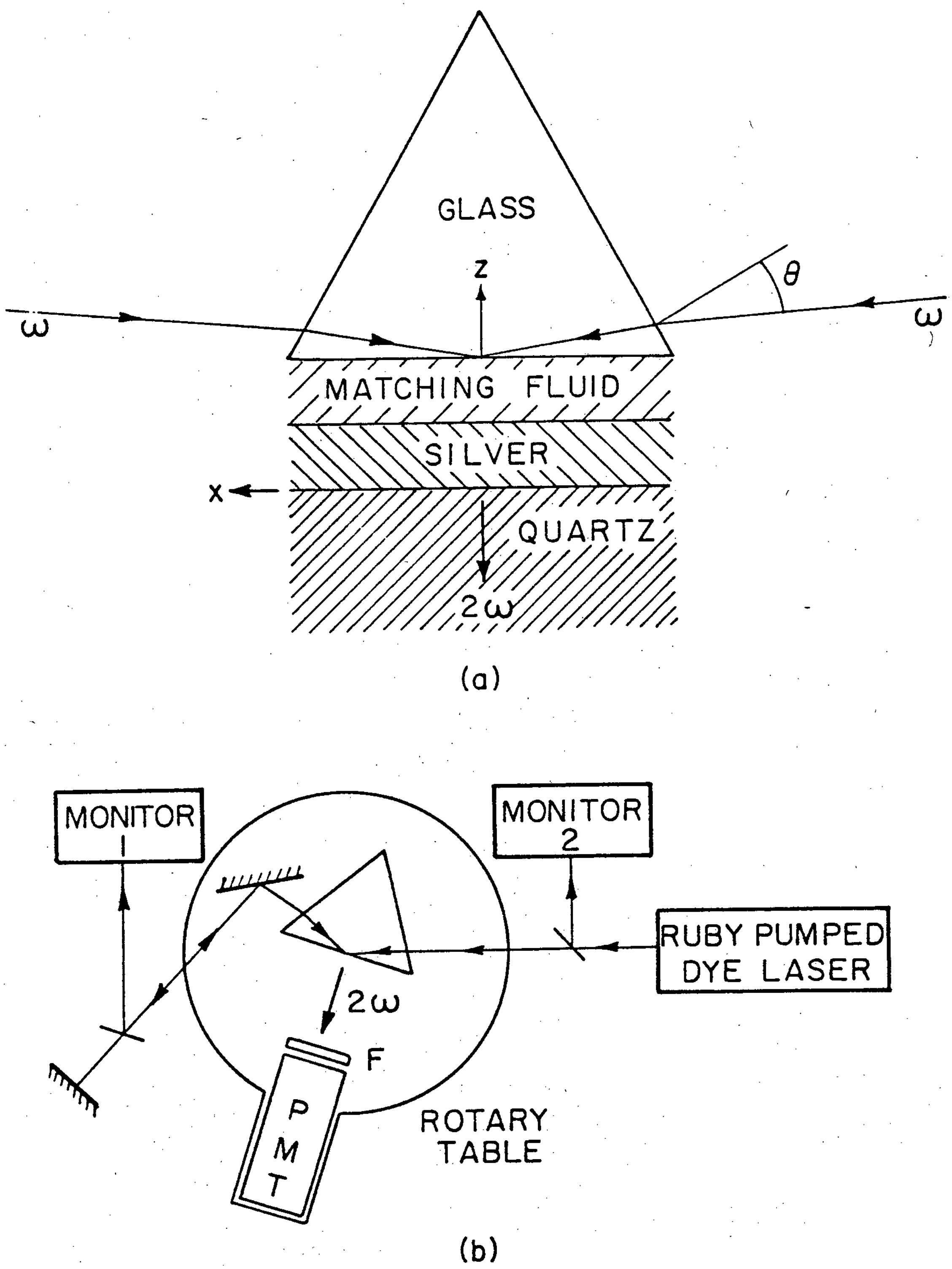
Figure Captions

Fig. 1(a) The sample assembly.

(b) Block diagram of the experimental set-up. F is a 10 cm long cell with saturated solution of $CuSO_4$ in water.

Fig. 2 Second harmonic intensity versus θ. θ is defined in Fig. 1(a).


GLASS
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SILVER
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QUARTZ
2ω
(a)
MONITOR 1
MONITOR 2
RUBY PUMPED DYE LASER
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F
P M T
ROTARY TABLE
(b)

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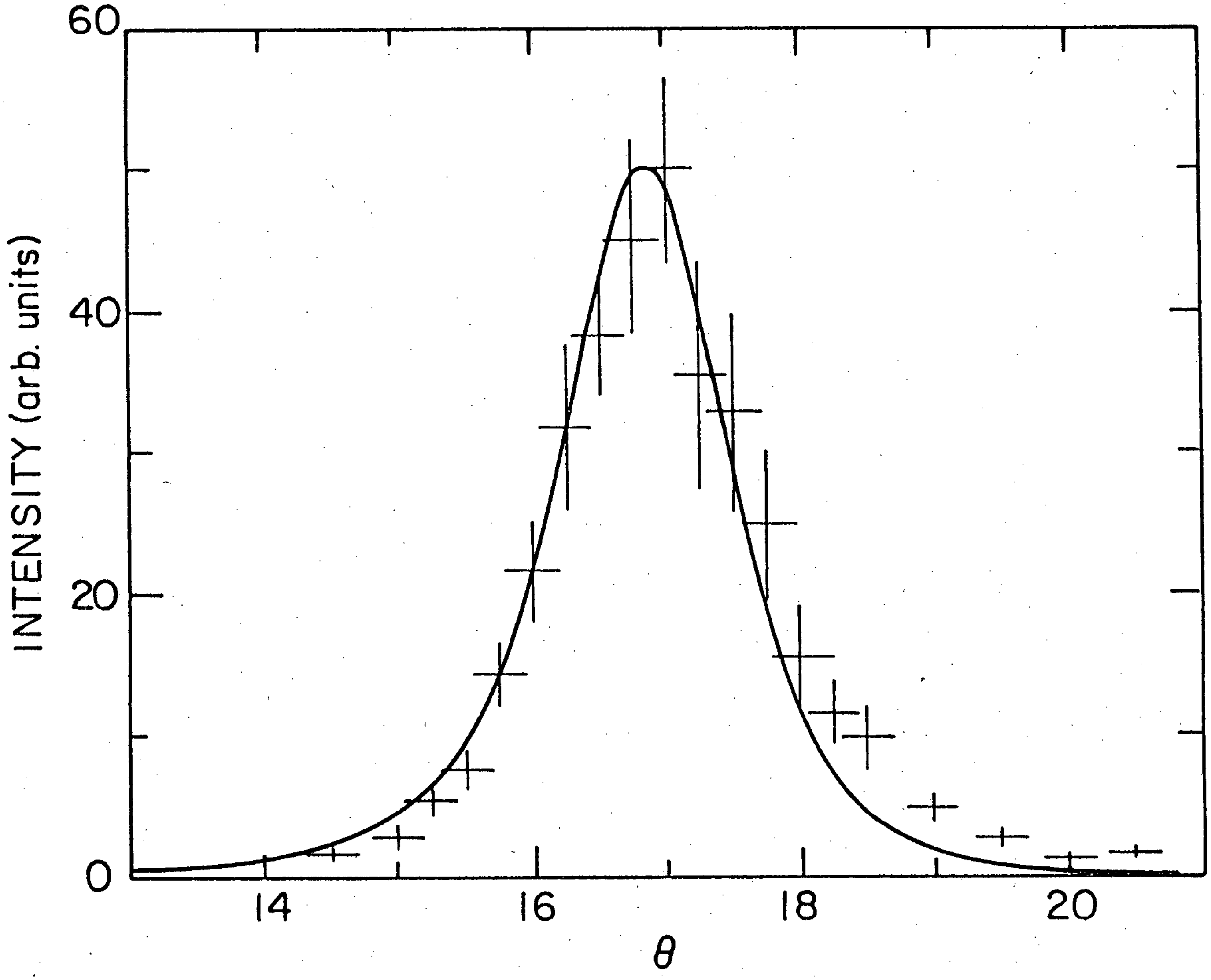

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