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SECOND HARMONIC AND SUM FREQUENCY GENERATION ON DYE-COATED SURFACES USING COLLINEAR AND NON- COLLINEAR EXCITATION GEOMETRIES

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ABSTRACT

Doubly resonantly enhanced sum frequency generation from rhodamine 6G monolayers adsorbed on glass substates is compared with resonantly enhanced second harmonic generation using a collinear excitation geometry. Second harmonic and sum frequency generation with a non-collinear excitation geometry is also reported where spatial filtering of the non-collinear output is shown to increase the scattered light rejection by more than 4 orders of magnitude.

SUMMARY

Second harmonic generation (SHG) with a collinear excitation geometry is becoming an important tool for surface studies.[1] Phase-matching on the surface is inherently satisfied since the nonlinear generation occurs over distances corresponding to only a few monolayers. We report resonantly enhanced sum frequency generation (SFG) for both collinear[2] and non-collinear[3] excitation geometries.

A pulsed dye laser probed the $S_1 \leftarrow S_0$ transition of rhodamine 6G. (ca. 525 nm) collinearly with the Nd:YAG fundamental (1064 nm) such that the sum frequency output was also resonant with the $S_2 \leftarrow S_0$ transition as shown in Fig. 1 which also shows the pumping scheme for resonantly enhanced SHG.

The results show that doubly resonant SFG is enhanced by more than 2 orders of magnitude relative to resonant SHG for submonolayer coverages of rhodamine 6G. In addition, the measured SFG excitation spectrum of rhodamine 6G is in good agreement with the calculated excitation spectrum shown in Fig. 2.

In a separate experiment, results for the non-collinear excitation of second harmonic and sum frequency generation from rhodamine 6G coated substrates demonstrate that the efficiency of nonlinear generation is not significantly reduced for crossing angles less than 15°. The output beam coherence can be exploited to achieve spatial separation from the incident beams, as shown

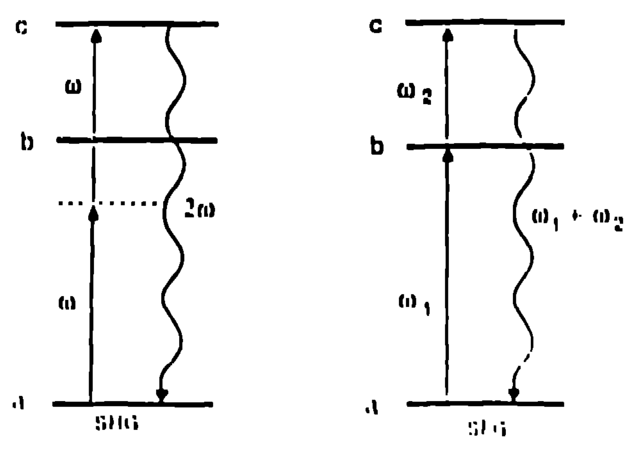


Fig. 1. Pumping schemes for resonantly enhanced SHG and SFG in rhodamine 6G.

for SHG and SFG in Fig. 3. Note that the noncollinear SHG output bisects the collinear outputs and that the relative signal intensities are in good agreement with the 2:1 ratio of noncollinear to collinear expected for equal input beam irradiances as shown in Fig. 3a. In Fig. 3b the noncollinear SFG output is resonantly enhanced angularly separated from the reflected visible beam by 2°.

The resulting spatial filtering of the nonlinear output possible with the noncollinear excitation geometry can increase the scattered light rejection by more than four orders of magnitude for crossing angles small as 6° as shown in Fig. 4 for SHG.

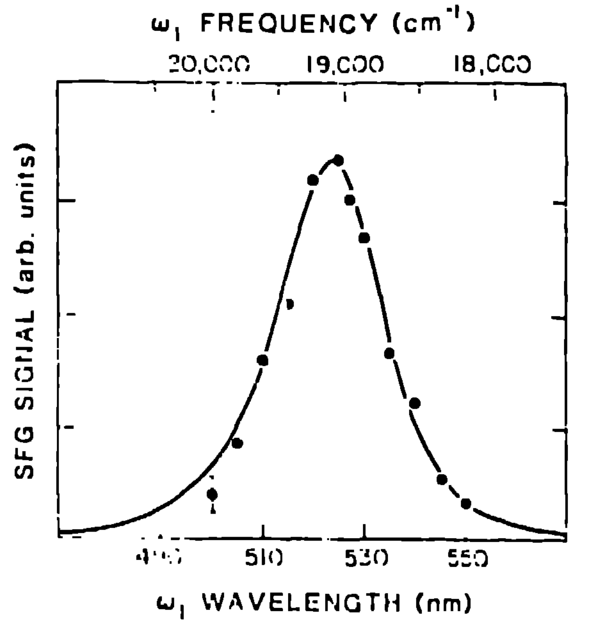


Fig. 2. SFG excitation spectrum, dots are the experimental data with the theory (solid curve) overlayed.

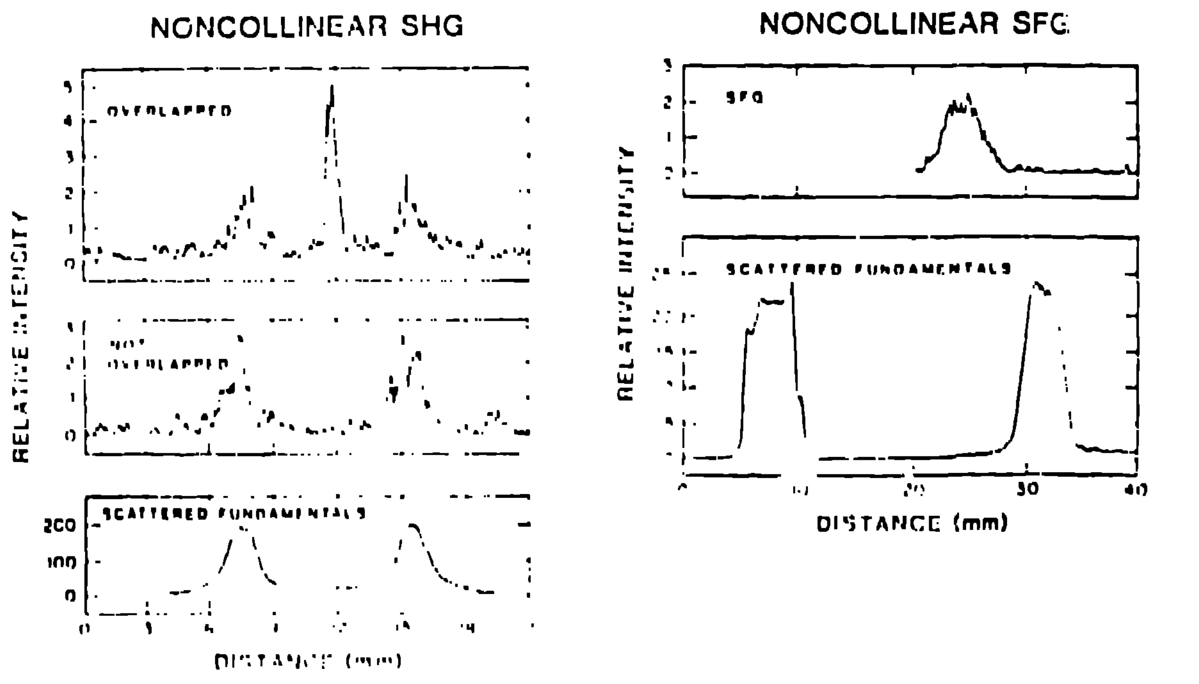
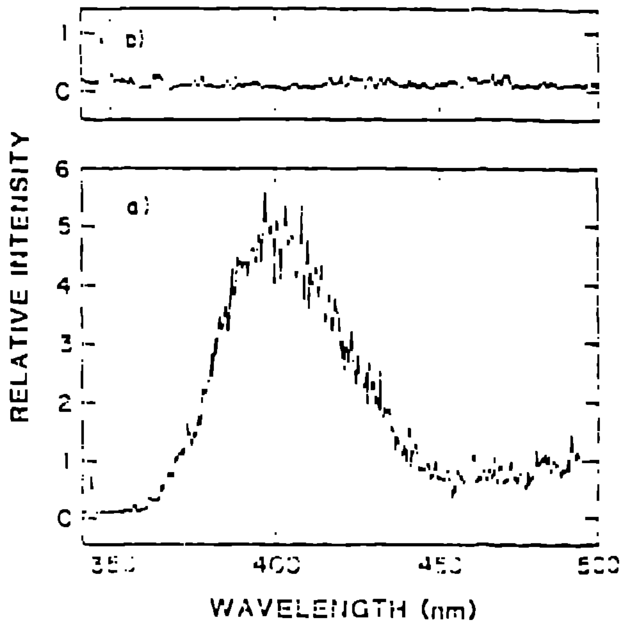


Fig. 3. a) Spatial location of the reflected input beams at 695 nm (bottom), SHG output at 347 nm for no input beam overlap on the surface (middle), and SHG output when the input beams are overlapped on the surface (top). The middle peak is the noncollinear SHG output. b) SFG output (top) at 351 nm with respect to the spatial location of the reflected input beams (bottom) at 1064 nm (left) and 525 nm (right).

Fig. 4. a) Monochromator scan showing collinear SHG output and the two-photon excited fluorescence of rhodamine 6G. The increasing background is due to the reflected fundamental at 695 nm. b) Same scan conditions except that the spatial filter passes only the noncollinear SHG ouput. The color filter used for collinear detection has also been removed.



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