

6. SUM-FREQUENCY GENERATION (SFG)

6.1. Principles of SFG

Shen, Berkeley (1987)

20 groups +

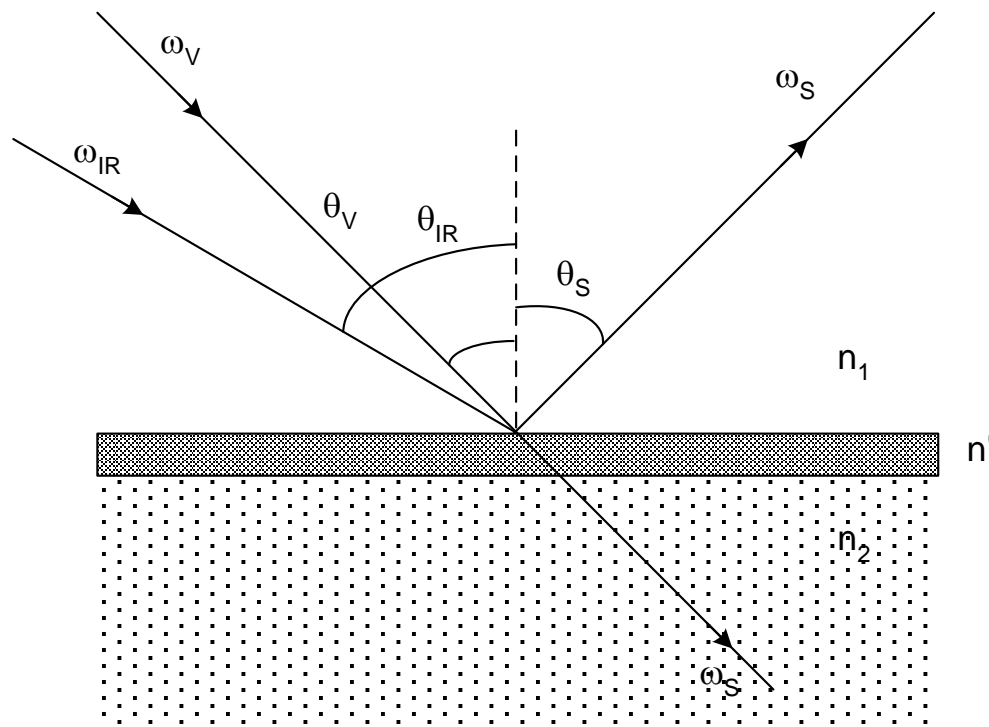
SFG a second order nonlinear optical process when ω_{IR} and ω_V two input lasers generate an output, ω_S

$$\omega_S = \omega_{IR} + \omega_V$$

It is forbidden for systems with inversion symmetry, but at surfaces the inversion centre is often broken.

When omegas (ω) give surface resonance, the SFG is resonantly enhanced.

Applicable to any interfaces accessible by light.



Geometry for sum –frequency generation

$$\theta_{\text{IR}} = 60^\circ$$

$$\theta_{\text{VIS}} = 45^\circ$$

IR-laser: 2.5-10 μm FELIX, free electron laser

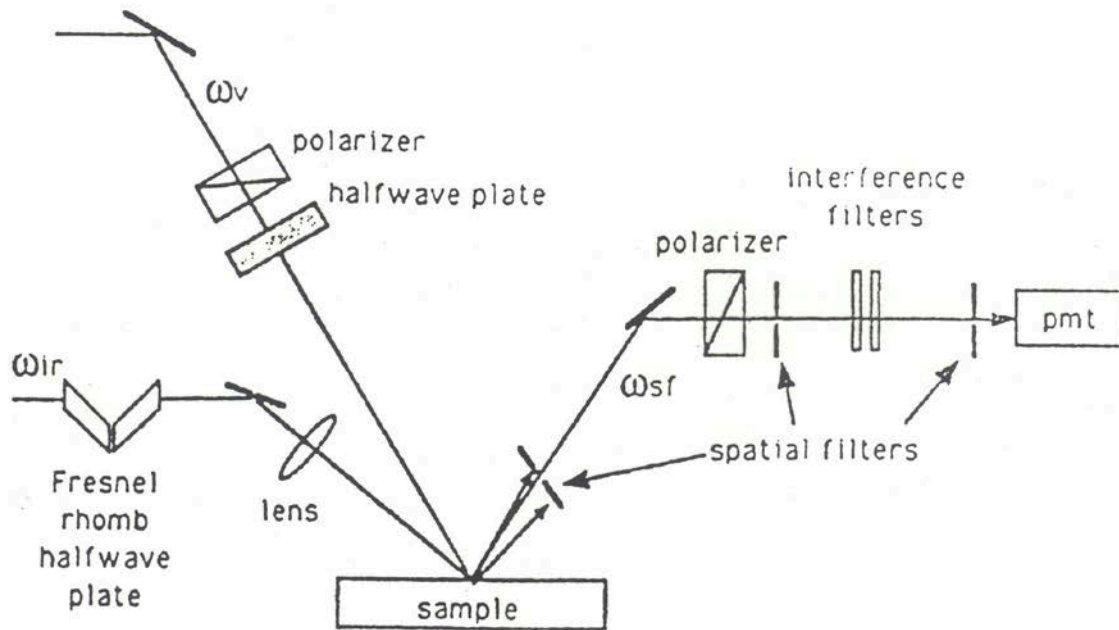
VIS-lasers: Nd:YLF (frequency doubled)

Nd:YAG (frequency doubled)

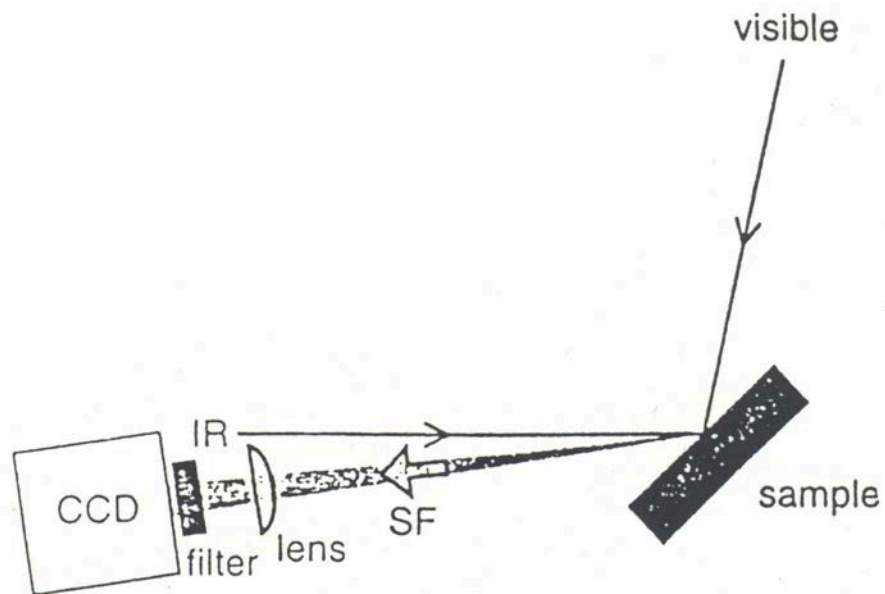
Impulses: microimpulse 5 μs

Macroimpulse 500 fs – 2 ps

Energy: 8-10 $\mu\text{J}/\text{pulse}$



Typical set-up for sum-frequency generation

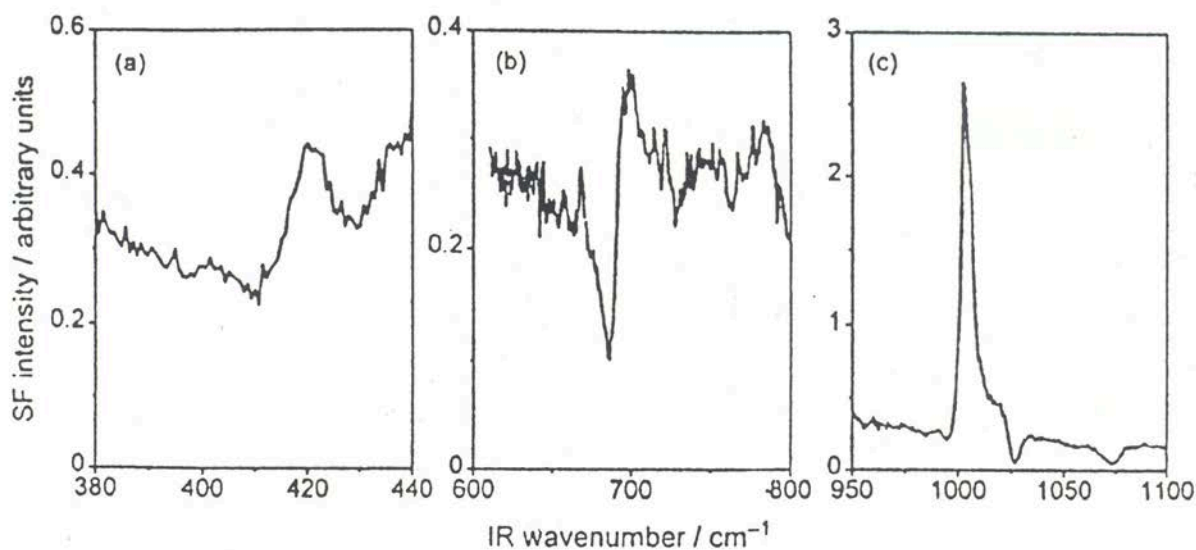


Another experimental arrangement for SFG spectroscopy (counter propagating geometry).

6.2. SOME APPLICATIONS OF SFG

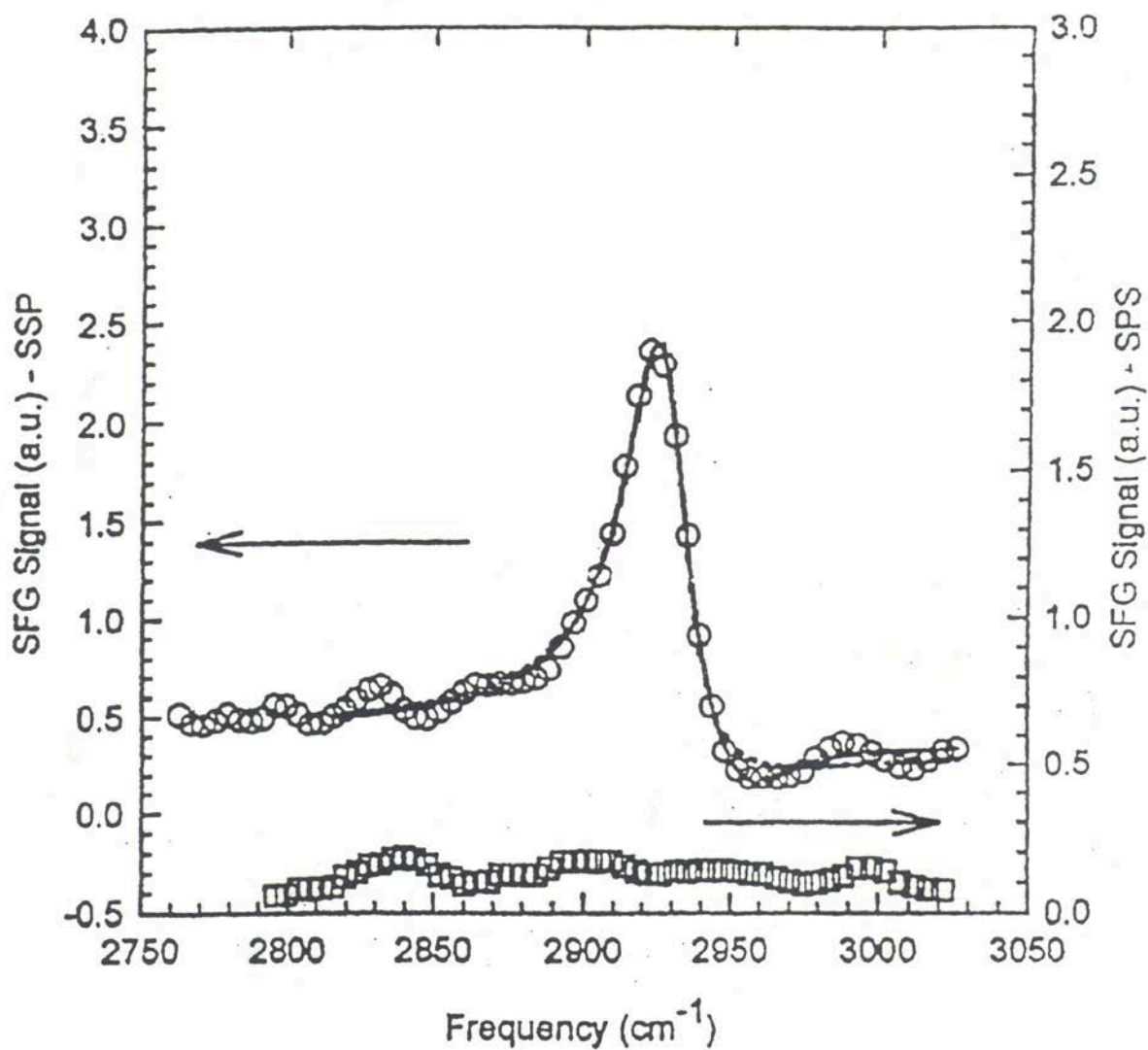
1. Solid/gas

Monolayer of thiophenole (TP) on Ag film (SERS, Auger, EELS, LEED studied before)



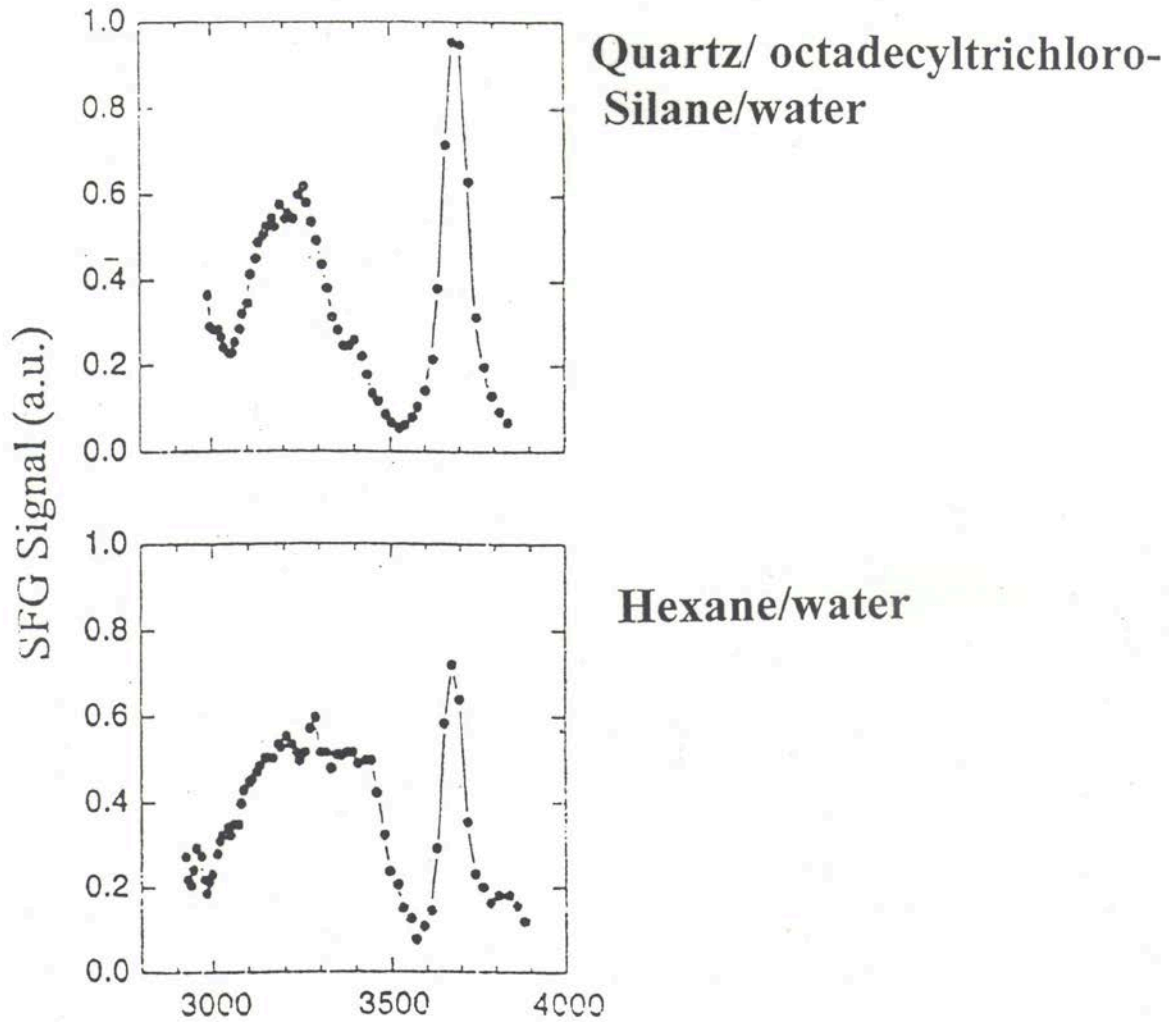
SFG spectra of Ag/TP showing resonances.

2. Liquid/vapour



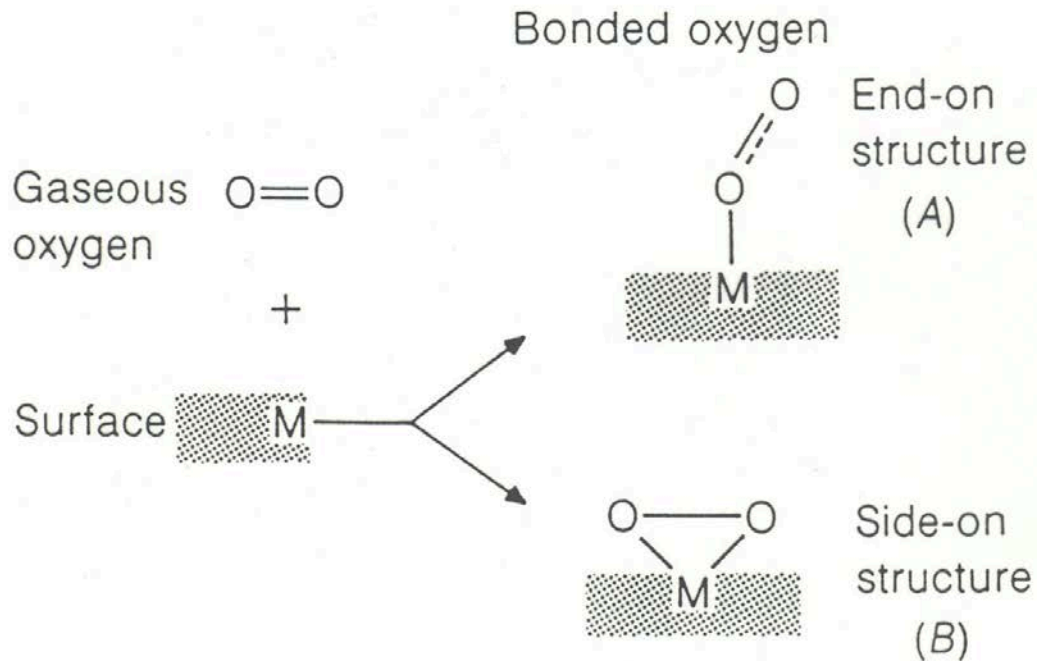
SFG spectra for the liquid/vapour interface of pure **acetone** with (\circ) **SSP** and (\square) **SPS** polarization combinations.

3. Solid/liquid:



SFG spectra of solid/liquid interface (SSP polarization)

CHEMISORBED O₂



Side-on (A) and end-on (B) structures of diatomic oxygen molecule adsorbed on metal surfaces at low temperatures.