

SMR/1238-22

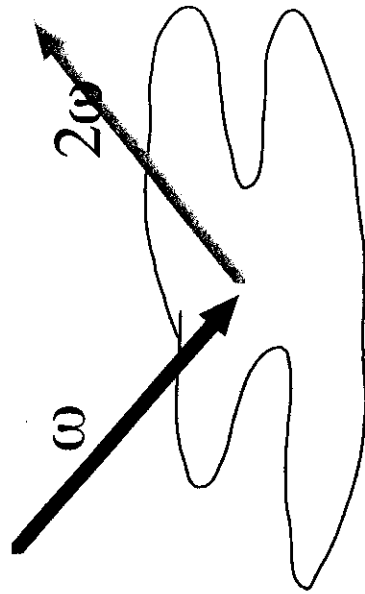
ADRIATICO RESEARCH CONFERENCE on
LASERS IN SURFACE SCIENCE

11–15 September 2000

Miramare – Trieste, Italy

*Optical second harmonic
microscopy of surfaces*

G. Mizutani
Japan Institute of Science and Technology
Ishikawa, Japan



Second Harmonic Microscopy serves
 a selective viewer of
 Asymmetry
 Surface/interface electronic levels

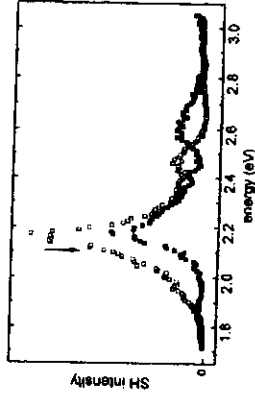


FIG. 1. SH spectrum of Cr_2O_3 at room temperature for wave vector parallel to the optical axis. Full and open squares refer to right and left circularly polarized light of the incoming light beam, respectively.

M. Fiebig, et al,

Appl. Phys. Lett. 66, 2906 ('95)

Nonlinear optical thin film resonators: design, fabrication, and applications. In: J. J. Burroughes, D. D. C. Bradley, A. N. Holmes, R. H. Friend, S. H. Tang, C. W. Tang, eds. *Physics and Chemistry of Organic Light-Emitting Diodes*. London: Chapman & Hall, 1997. p. 1-10.

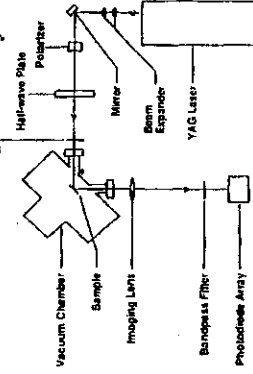


FIG. 2. Schematic diagram of the experimental setup.

K. A. Schultz et al,
 J. Chem. Phys. 97,
 6958 ('92)

Other examples : ferroelectric materials, surface/
 interface magnetism, LB films, rat tail tendon

Oxidation of CO on Pt(110)

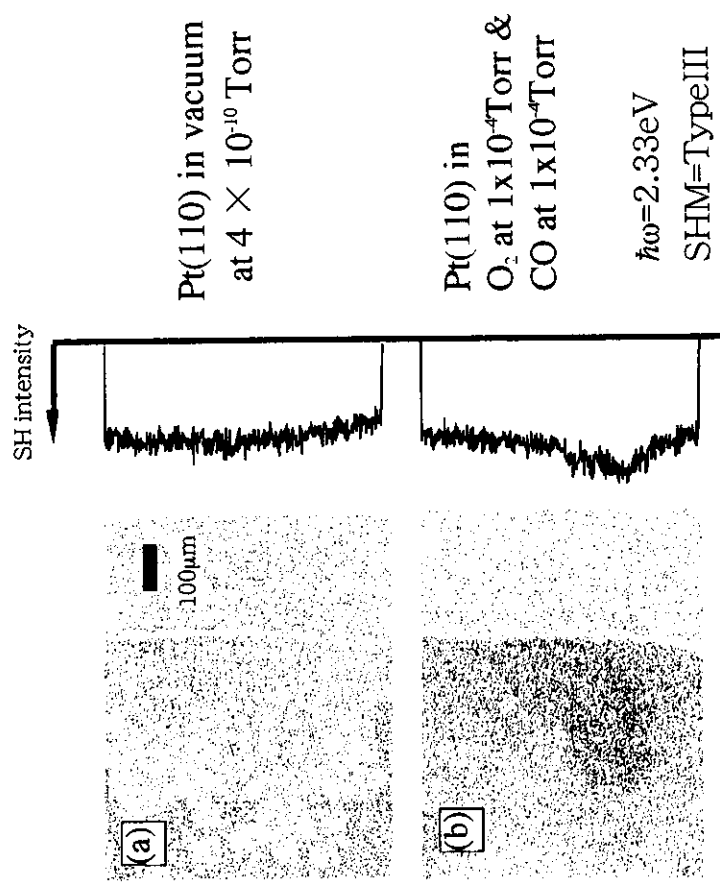
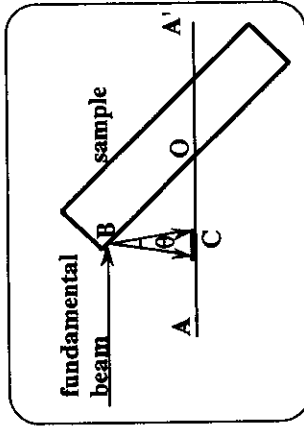
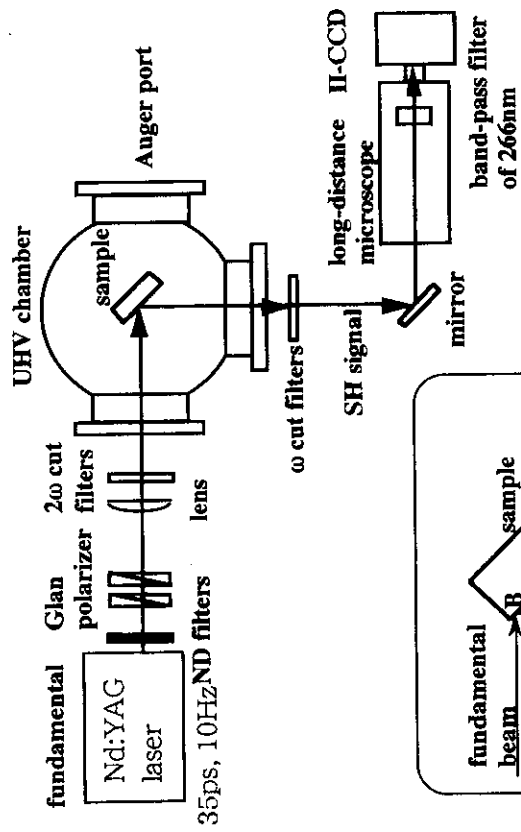
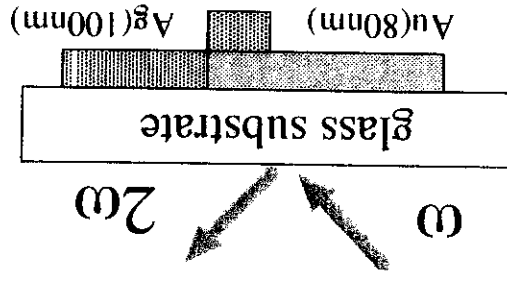
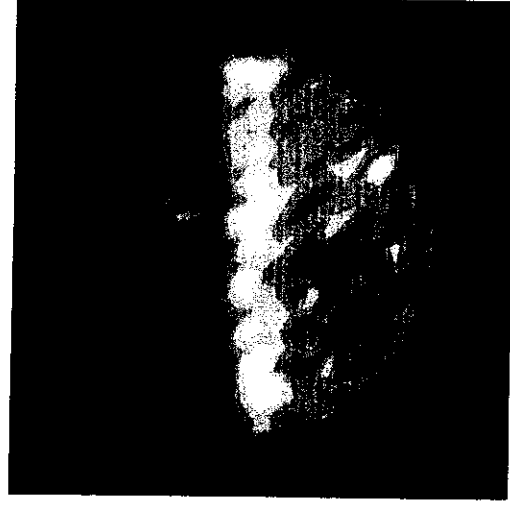


Fig. 2 (a) SH intensity image of Pt(110). Dark dots are detected photons and the sample is seen on the left. (b) SH intensity image of Pt(110) under stationary reaction conditions at 298K with $P_{O_2} = P_{CO} = 1 \times 10^{-4}$ Torr. The accumulation time was 300min. The curves on the right are one-dimensional profiles of the SH intensity.

SH Intensity patterns of metal stacks



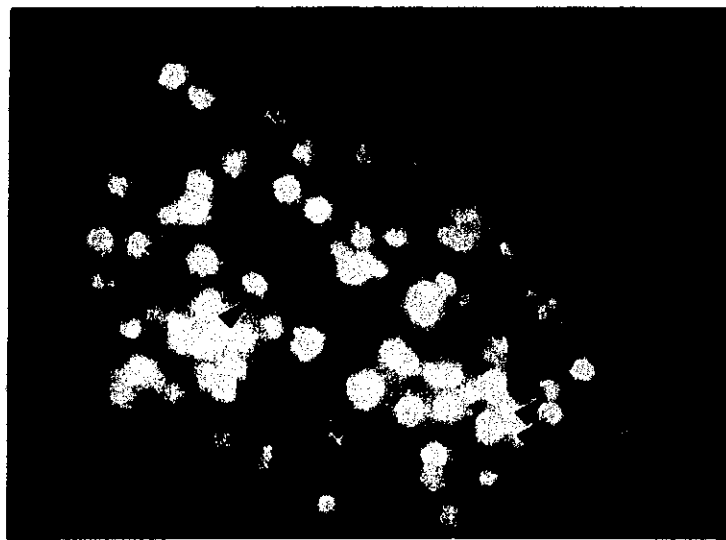
SHM=Type I
 $\hbar\omega=1.17\text{eV}$

→ No enhancement by roughness
 cf. G. T. Boyd et al. Phys. Rev. B30, 519 (1984)

indicate origins of the enhancement
 suppression of SH intensity in the
 70-layered region

- × SH response from the buried interface
- × Interdiffusion of metal atoms
- ? Difference in surface roughness
 =Difference in SPP excitation
- ? Contact potential difference

SH intensity image



SHM=Type II
 $\hbar\omega=1.17\text{eV}$

Linear image



The structure of a starch granule

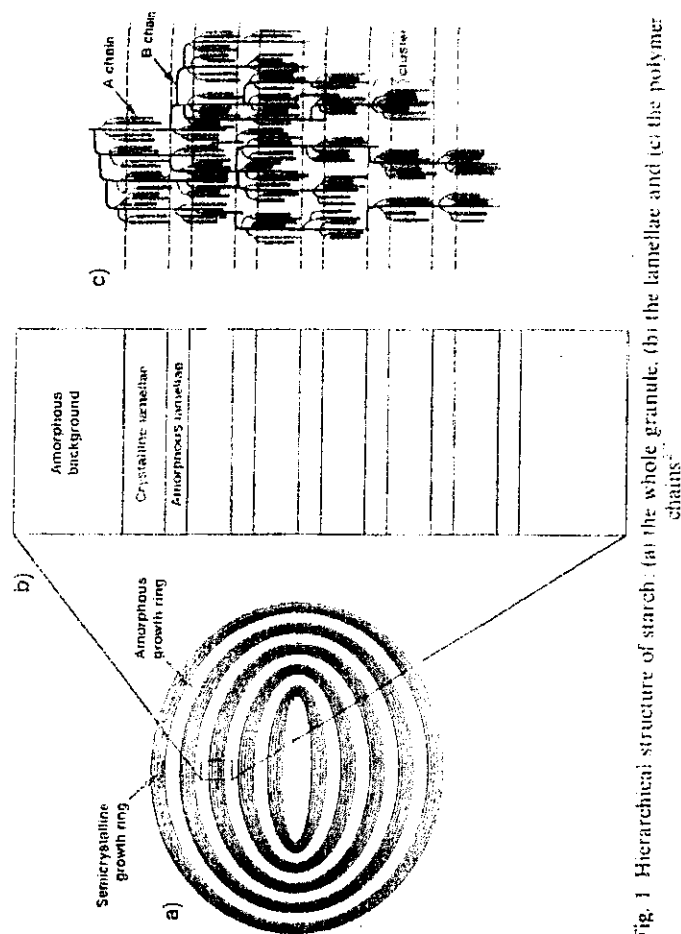


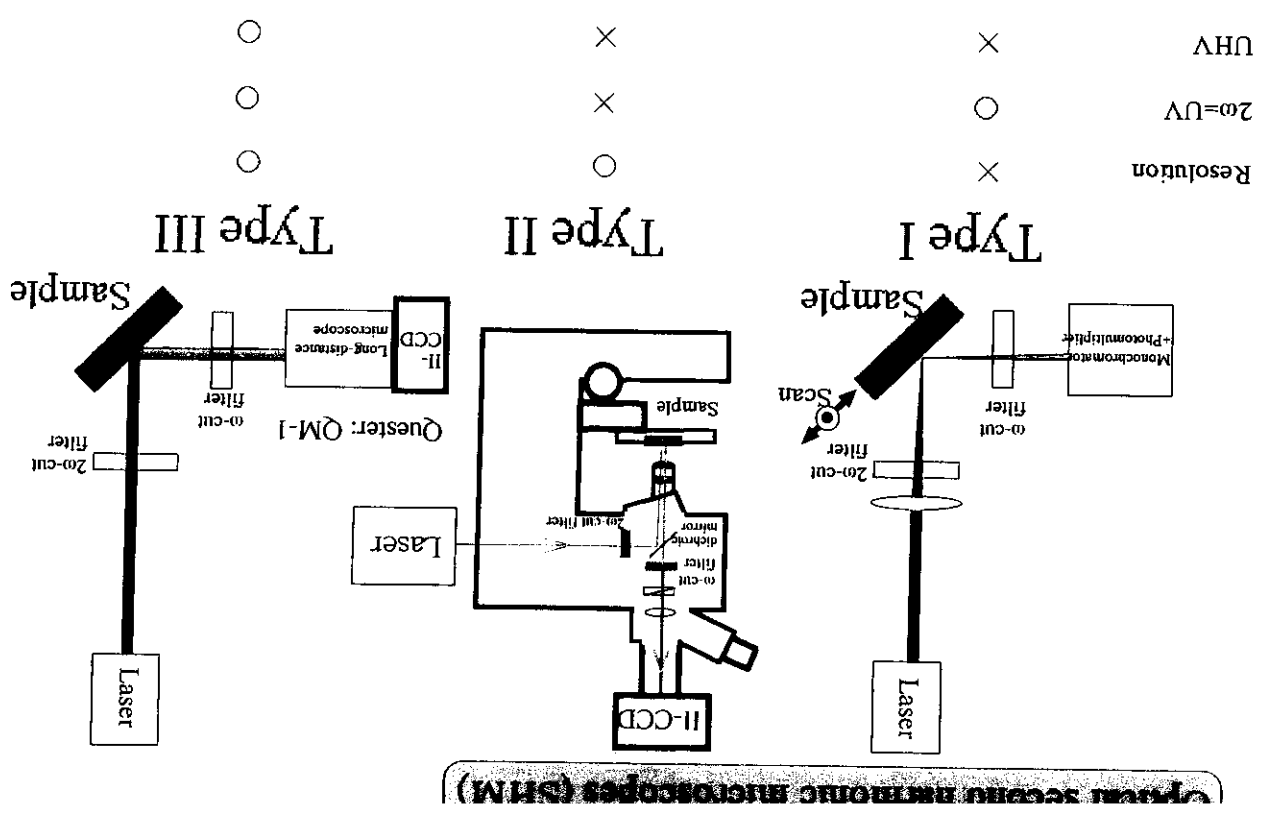
Fig. 1 Hierarchical structure of starch: (a) the whole granule, (b) the lamellae and (c) the polymer chains.

T. A. Waigh et al, Faraday Discuss., 1996, 103, 325

- The double helices are arranged into a crystalline cluster unit.
- The structure is very similar to that of quartz.

Kare Larsson, Acta chemica Scandinavica 45 (1991) 840

- I will show SH intensity images of
1. Oxidation of CO on Pt(110)
 2. Multi-layered metal stacks
 3. Starch granules in plants



Origin of the local enhancement of SHG

Enhancement of SHG

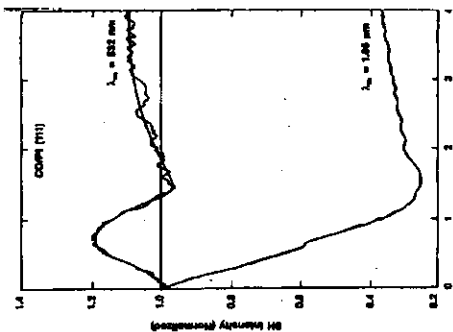


Figure 7. The second-harmonic response from Pt(111) as a function of exposure to CO for two different incident wavelengths. Both curves are normalized to the clean surface SHG response for each laser wavelength.

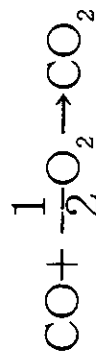
SHG from CO/Pt(111)
Grubb et al.
J. Phys. Chem. 92, 1419 (1988)

Spatial inhomogeneity



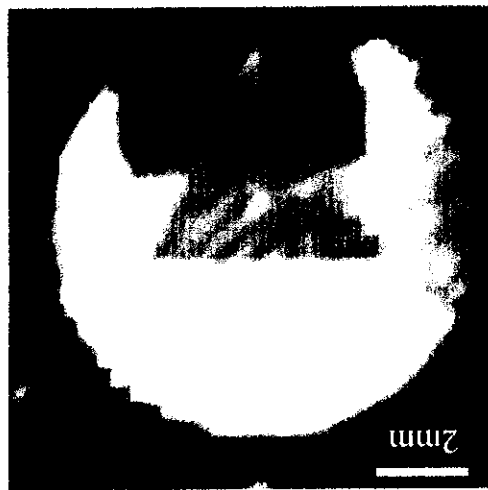
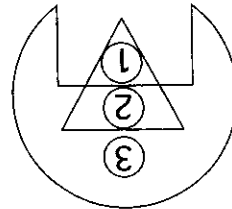
110μm

PEEM observation
of CO+O₂/Pt(110)



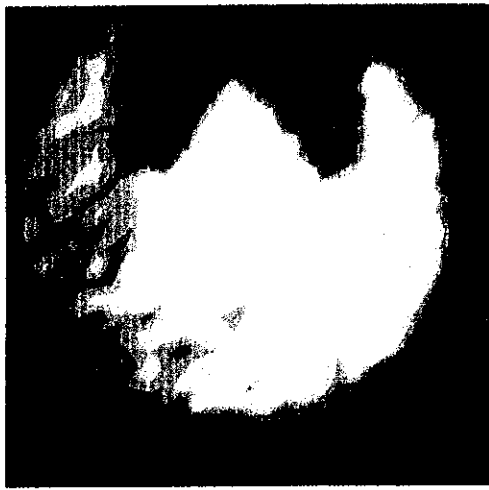
Intense SHG → adsorbed CO

SH Intensity patterns of metal stacks

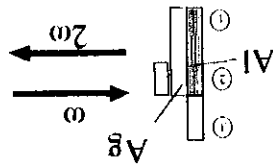
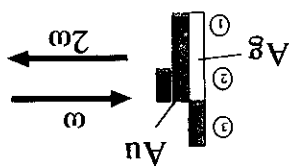


- ① Ag(30nm) on Al(30nm)
- ② Ag(60nm) on Al(30nm)
- ③ Ag(30nm)

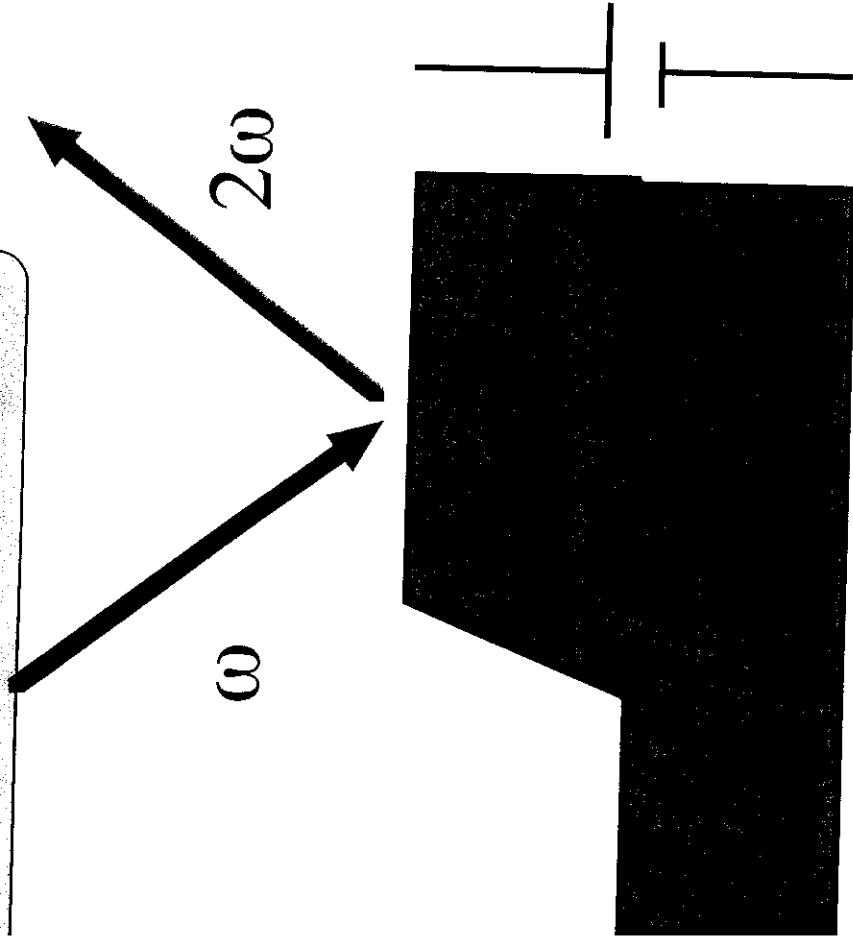
SHM=Type I
 $\hbar\omega=1.17\text{eV}$



- ① Au(10nm) on Ag(50nm)
- ② Au(40nm) on Ag(50nm)
- ③ Au(30nm)



Interpretation of the SH intensity distribution



we see the contact potential
reference distribution
Correlation was checked for
Ag/Au, Au/Ag, Ag/Cu, Ag/Al

Chara corallina

SHM=Type II
 $h\nu=1.17\text{eV}$

SH intensity image



Linear image

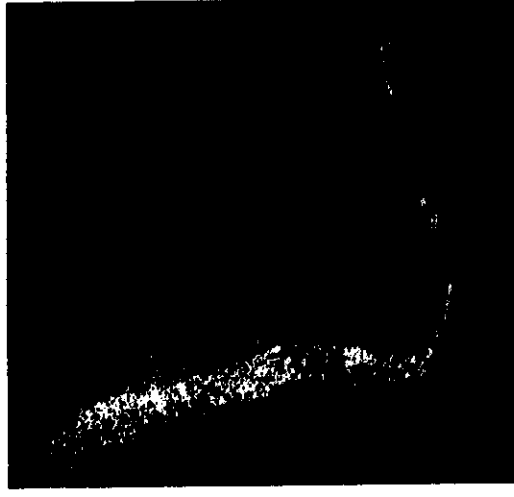


○Ultra-high vacuum optical second harmonic microscope (UHVSHM) giving 2D images has been constructed.

○Metal stacks give interesting SH intensity images. Origins of the contrast of SH intensity still require some discussion.

○We have observed SH intensity images of plants for the first time. Starch granules are observed selectively.

SH intensity image



Linear image



Cellulose