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ADRIATICO RESEARCH CONFERENCE on

LASERS IN SURFACE SCIENCE

11-15 September 2000

Miramare - Trieste, Italy

Coherent and incoherent dynamics of electronic surface states of Si(111) and Si(001)

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Motivation

Why Study Surface Carrier Dynamics?

- Model for dynamics at interfaces
- Surface photochemistry
  - relaxation rate of photoexited carriers
    → yield of photochemical reactions
- Performance of devices
  - accurate fundamental carrier relaxation rates
    → correct modeling of state-of-the-art Si devices

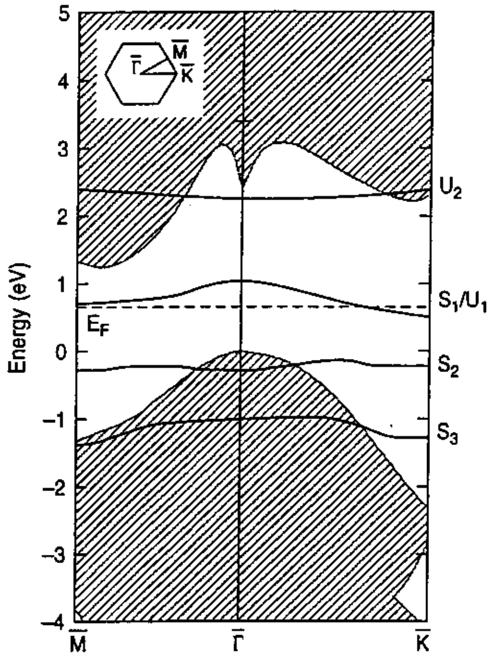
Why Probe Relaxation Times with Nonlinear Optical Techniques?

- Genuine surface signal for $\chi^{(2)}$, $\chi^{(4)}$
- Applicable at high excitation conditions
- Applicable to many different interfaces

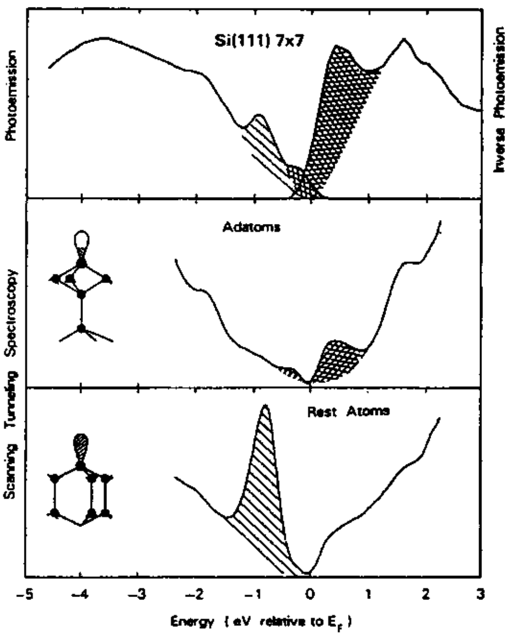
This work:

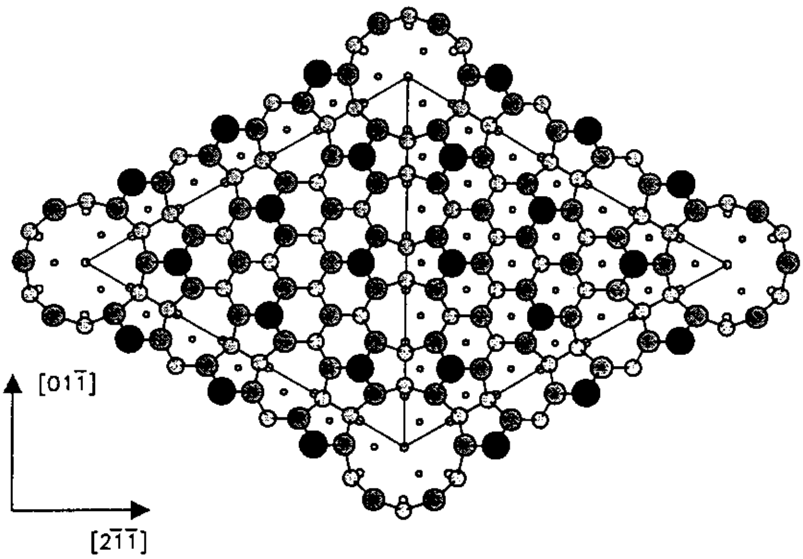
- Combination of SHG and DFWM
  → coherent technique to study carrier scattering processes at surfaces
- Opportunity of learning about some of the most fundamental quantum mechanical processes in surfaces

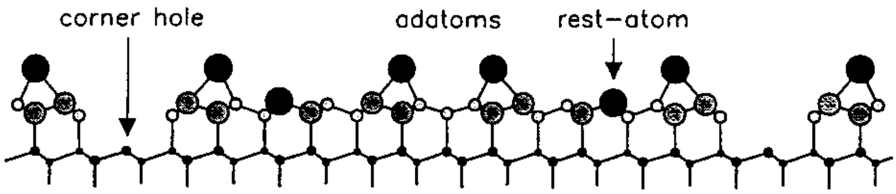
Si(111)7×7



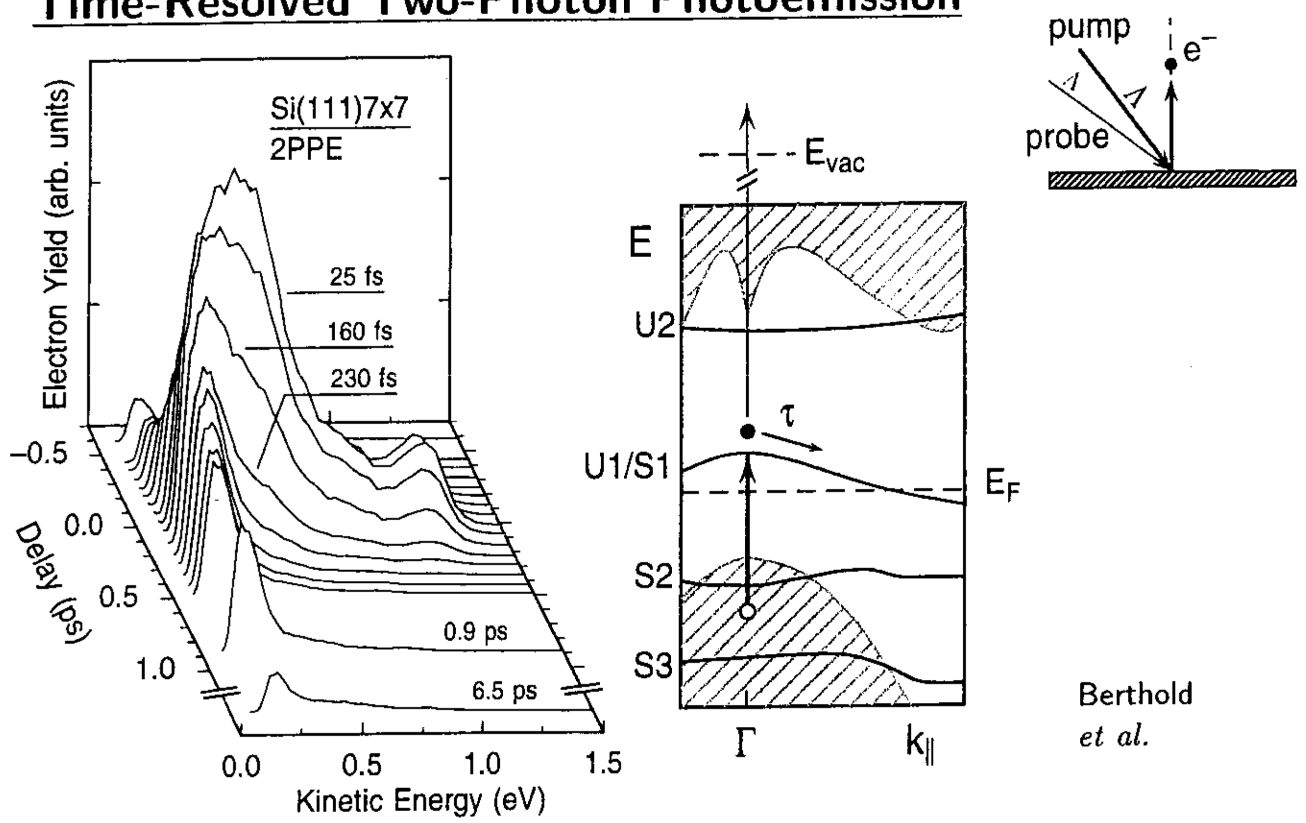
F.J. Himpsel





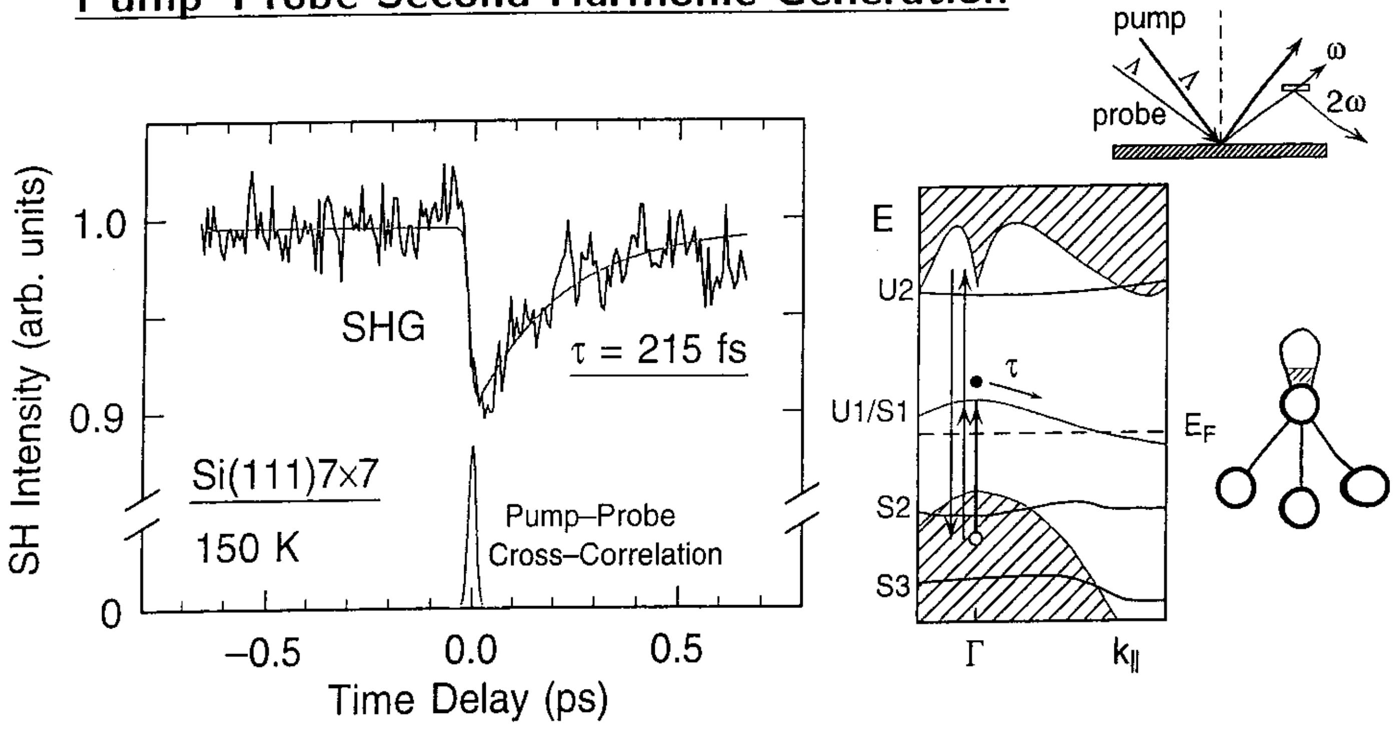


Time-Resolved Two-Photon Photoemission



Berthold *et al.*

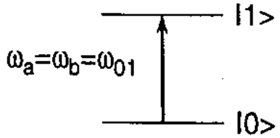
Pump–Probe Second-Harmonic Generation



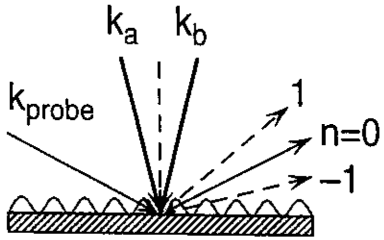
phys. stat. sol. (a) **175**, 169 (1999).

Diffraction from Transient Gratings

Grating formation:



P: Polarization
N_{01}: Population difference



$\chi_s^{(2)}(x) = \chi_s^{(2)}(x+l_g)$

$$\frac{\partial}{\partial t}P_j^{(1)} = \frac{i\mu}{\hbar}E_j - \frac{1}{T_2}P_j^{(1)}, \qquad j = \mathrm{a,b}$$

$$\frac{\partial}{\partial t}N_{01}^{(2)} = \frac{i\mu}{\hbar}(E_\mathrm{a}P_\mathrm{b}^{*(1)} - P_\mathrm{a}^{(1)}E_\mathrm{b}^*) + \mathrm{c.c.} - \frac{1}{T_1}N_{01}^{(2)}$$

$$\frac{\partial}{\partial t}P_\mathrm{d}^{(3)} = -2\frac{i\mu}{\hbar}E_\mathrm{p}N_{01}^{(2)} - \frac{1}{T_2}P_\mathrm{d}^{(3)}$$

Information from diffracted signal (limiting cases):

a) delay $\tau_\mathrm{b} = 0, \tau_\mathrm{probe} > 0$
$\Longrightarrow$ Population relaxation T_1 (& diffusivity)

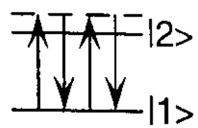
b) delay $\tau_\mathrm{b} > 0, \tau_\mathrm{probe} = \tau_\mathrm{b}$
$\Longrightarrow$ Dephasing T_2

self-diffraction: $\mathbf{k}_\mathrm{probe} \equiv \mathbf{k}_\mathrm{b}$

DFWM vs. SH-Diffraction

Four-wave mixing

$\chi^{(3)}$



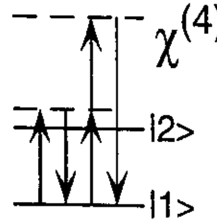
- bulk (& surface) sensitive

$$\mathbf{P}^{(3)}(\mathbf{K}_\mathrm{d}^{(\pm1)};\omega) = \chi^{(3)} : \mathbf{E}_\mathrm{a}^*(\mathbf{k}_\mathrm{a})\mathbf{E}_\mathrm{b}^2(\mathbf{k}_\mathrm{b})$$

$$K_{\mathrm{d},x}^{(\pm1)} = k_{\mathrm{b},x} \pm (k_{\mathrm{b},x} - k_{\mathrm{a},x})$$

2ω diffraction (five-wave mixing)

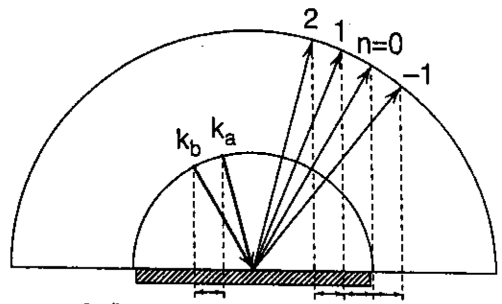
$\chi^{(4)}$



- surface sensitive

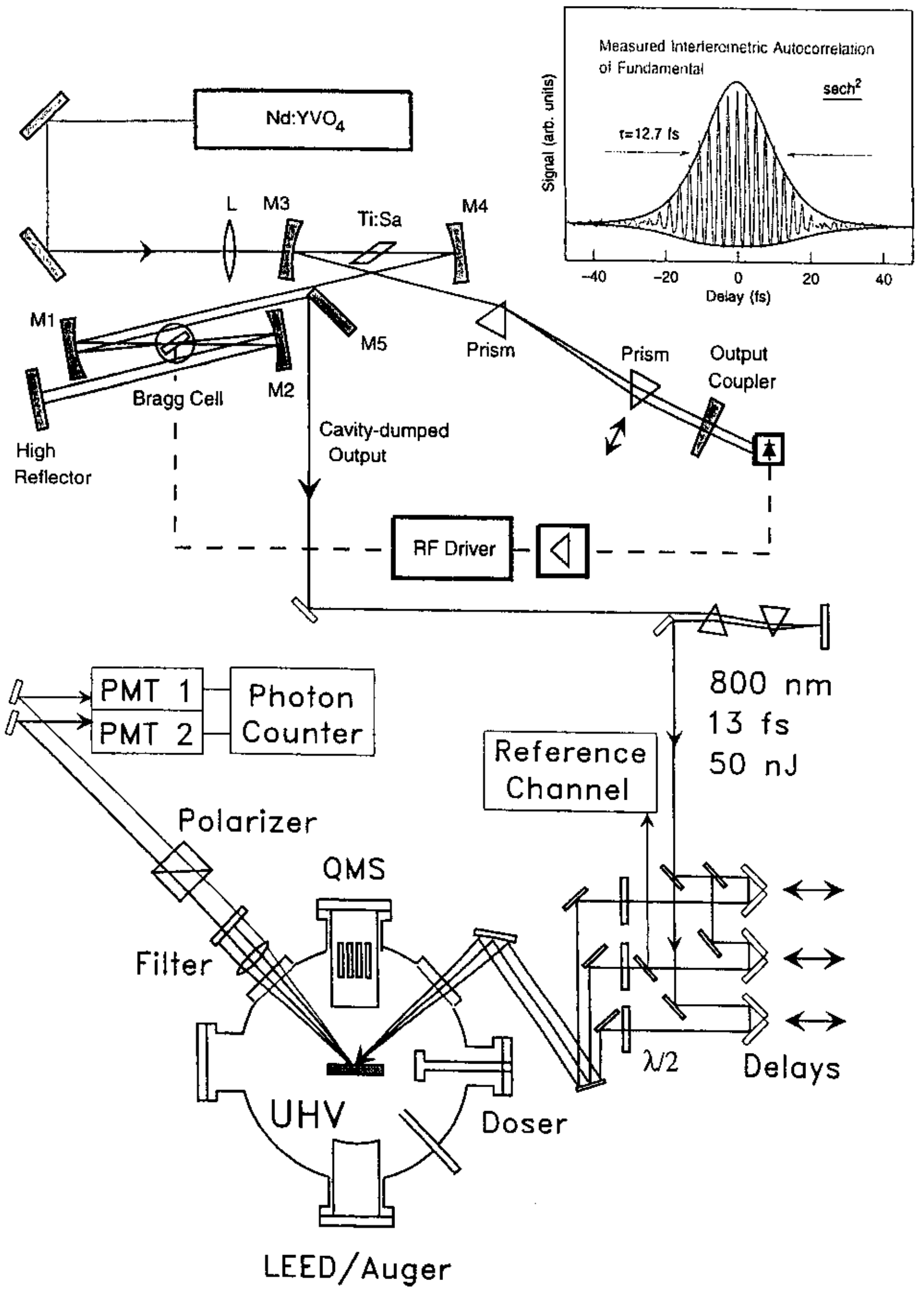
$$\mathbf{P}^{(4)}(\mathbf{K}_\mathrm{d}^{(\pm1)};2\omega) = \chi^{(4)} : \mathbf{E}_\mathrm{a}^*(\mathbf{k}_\mathrm{a})\mathbf{E}_\mathrm{b}^3(\mathbf{k}_\mathrm{b})$$

$$K_{\mathrm{d},x}^{(\pm1)} = 2k_{\mathrm{b},x} \pm (k_{\mathrm{b},x} - k_{\mathrm{a},x})$$



$2\pi/l_g = k_{b,x} - k_{a,x}$

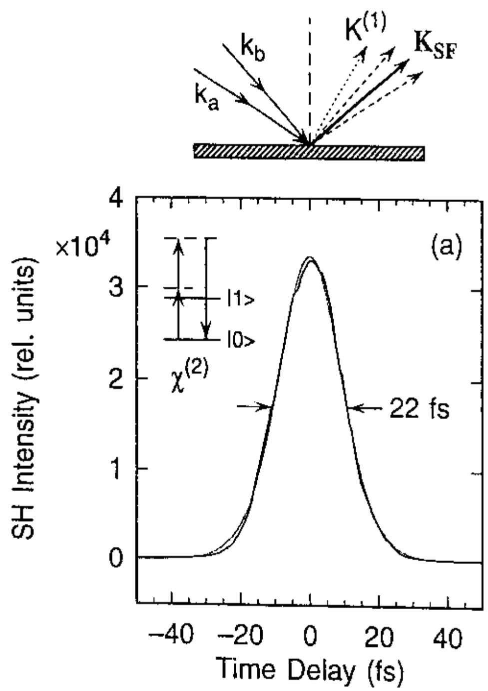
Experimental Setup



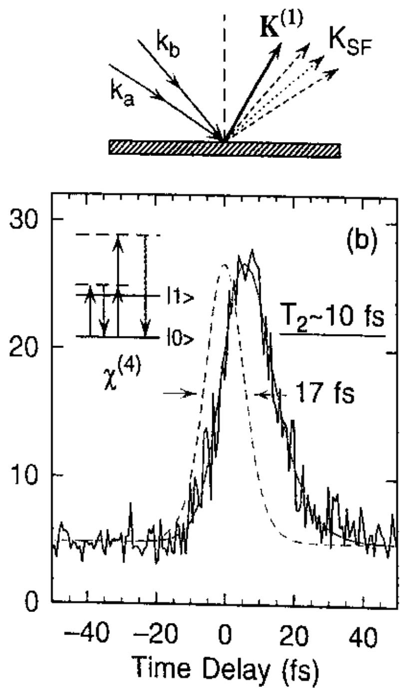
Experimental Data

Si(111)7×7, 80 K

Sum-Frequency Signal



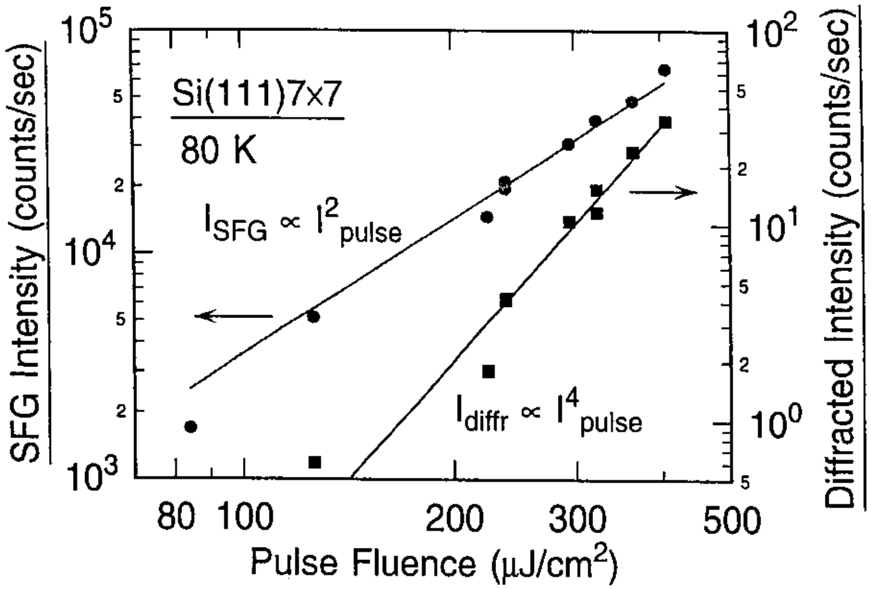
Diffracted Signal



⇒ Pulse duration:
$\tau_p < 14$ fs
(on sample in UHV)

⇒ Dephasing time:
$T_2 \simeq 10$ fs

Pulse Fluence Dependence



Sum-frequency signal

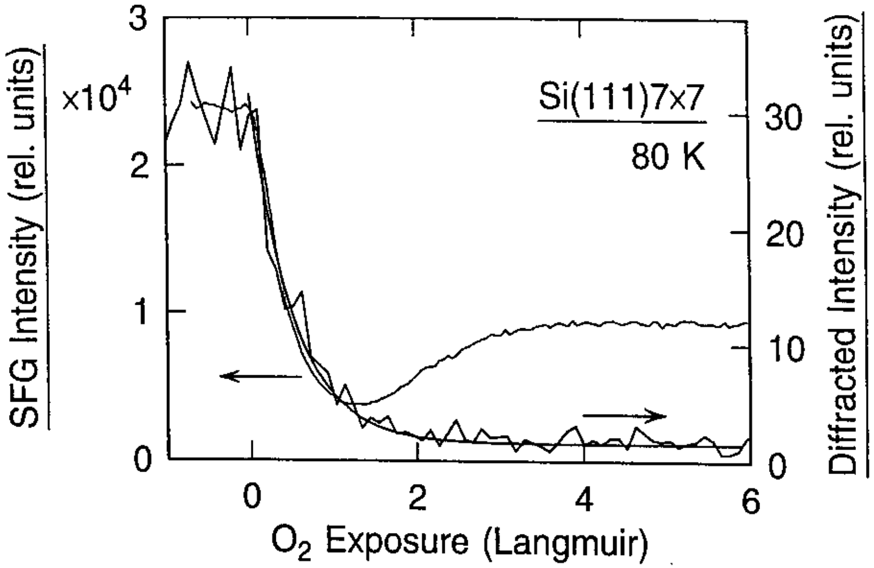
$$\mathbf{P}_s^{(2)}(\mathbf{K}_{SF}; 2\omega) = \chi_s^{(2)} : \mathbf{E}_a(\mathbf{k}_a)\mathbf{E}_b(\mathbf{k}_b)$$

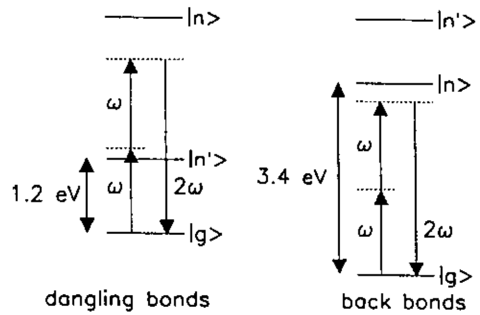
Self-diffracted signal

$$\mathbf{P}_s^{(4)}(\mathbf{K}_d^{(+1)}; 2\omega) = \chi_s^{(4)} : \mathbf{E}_a^*(\mathbf{k}_a)\mathbf{E}_b^3(\mathbf{k}_b)$$

$$I_d(2\omega) \propto |\mathbf{P}_s^{(4)}(2\omega)|^2$$

Coverage Dependence

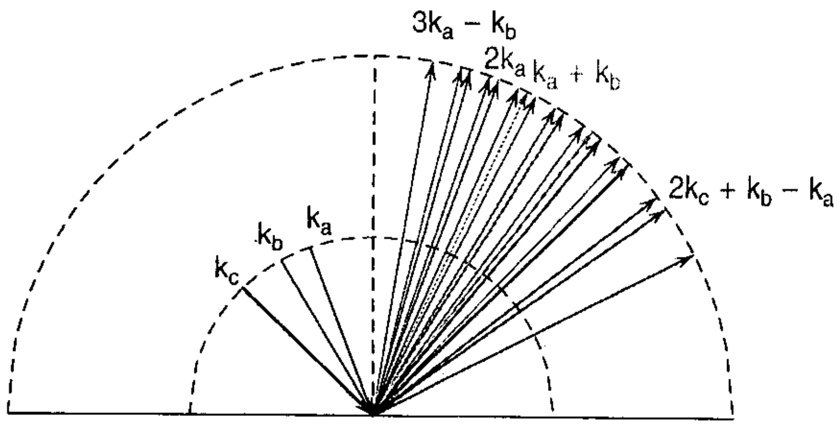


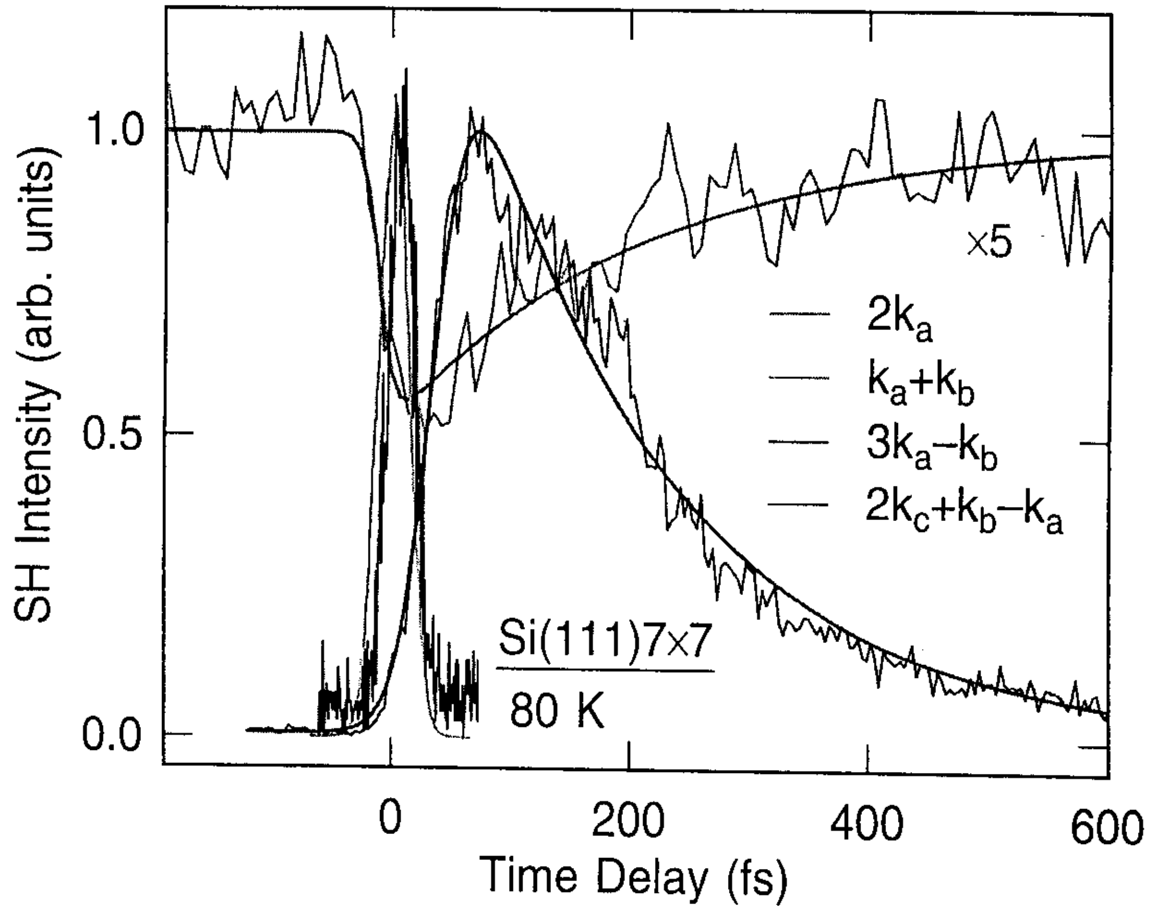


Appl. Phys. B **68**, 383 (1999).

- minimum in the SFG signal due to destructive interference between dangling bond and back bond terms in $\chi_s^{(2)}$
- no minimum in diffracted SHG signal: the only contribution is dangling bond term $\chi_{db}^{(4)}$

SH Signals into Various Directions





Numerical Simulation

- density-matrix formalism:
  semi-classical approach
  - electromagnetic light field treated classically
  - material system treated quantum-mechanically by its density matrix ρ
  - light–matter interaction Hamiltonian in the electric-dipole approximation
- Liouville equation
  $\frac{\mathrm{d}}{\mathrm{d}t}\rho = \frac{1}{i\hbar}[H, \rho] + \frac{\mathrm{d}}{\mathrm{d}t}\rho_{\mathrm{relax}}$
- numerical integration of Bloch equations for $\chi^{(4)}$ second-harmonic diffraction
- for independent three-level system with detuning
- to 4th order in terms of the electric field E
- relaxation-time (Markovian) approximation
- summation over range of both detunings to simulate inhomogeneous broadening
- slowly-varying-envelope approximation for sech^2 or Gaussian pulses

Conclusion

- Demonstration of surface sensitive transient grating experiment ($\chi_s^{(4)}$ process)
  - 2-beam self-diffraction geometry
  - 3-beam diffraction geometry
- Excellent time resolution due to cavity-dumped Ti:sapphire laser (pulse duration in UHV chamber < 14 fs)

Pump-Probe Relaxation Times	Dephasing Times
• T_1 **time: population relaxation**	• T_2 **time: polarization relaxation**
• origin: multiple scattering of excited electrons out of optically coupled region in k-space	• origin: single, phase-destroying scattering processes $\rightarrow$ eventual loss of coherence
• $T_1 = 100$ fs – 2 ps	• $T_2 \simeq 10$ fs (inhom. broadening)