Interferometric third-harmonic generation in thin optical films using ultrashort laser pulses

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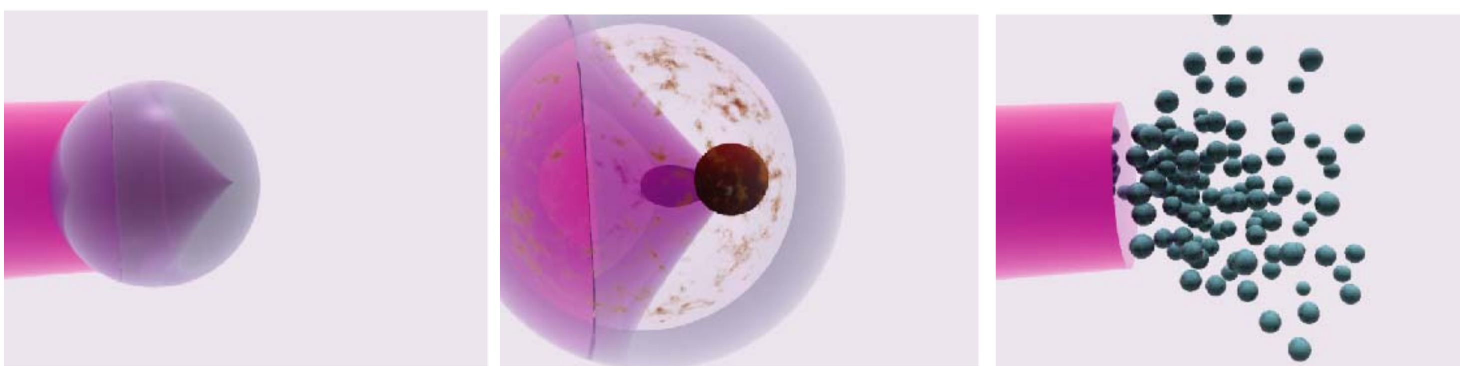
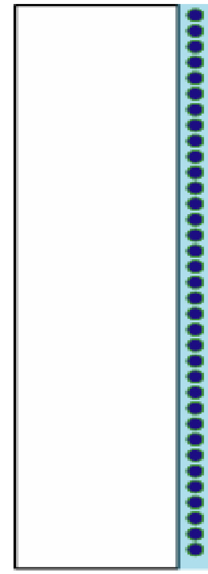
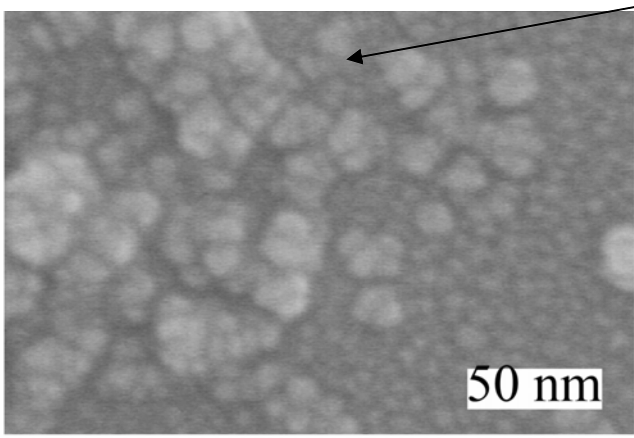


Ultrafast THG is an interface probe

- Measure $\chi^{(3)}$ of nonlinear thin films (e.g. Nano- AlN/GaN or biological)
- Must eliminate the substrate (background)
- Or, include the substrate (phase-sensitivity)
- Femtosecond lasers have unique interface sensitivity





<u>Structural details are available through NLO</u>:

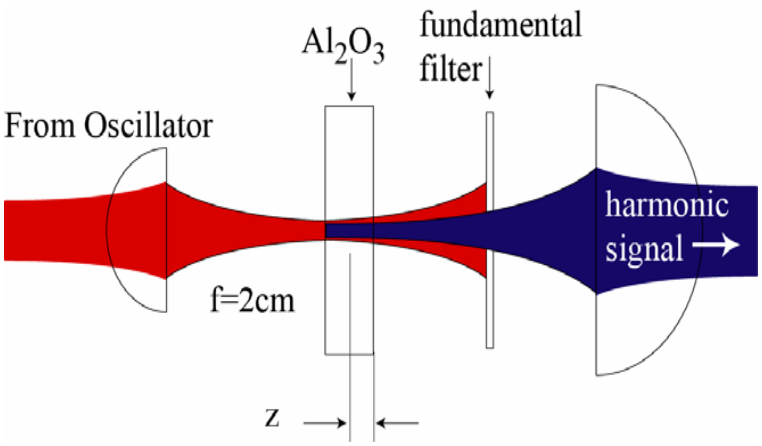
- SHG->Inversion Asymmetry
- THG->discontinuities in bulk parameters
- index, group velocity, etc.

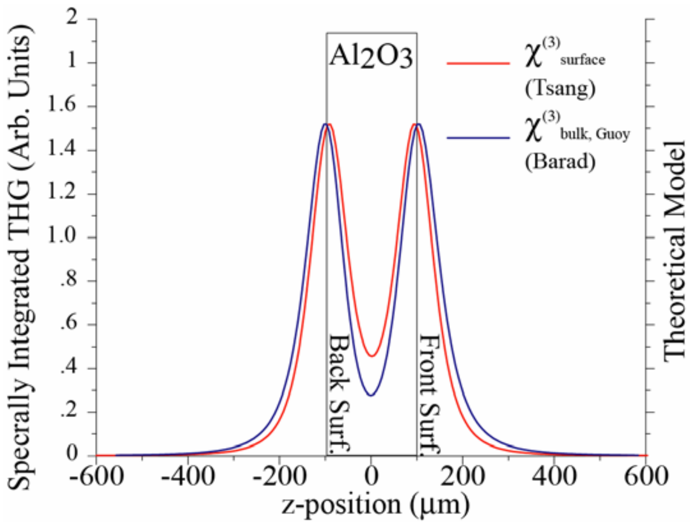
Early models of interface THG

- (Z-Scan)
  - T.Tsang, et.al. *Phys. Rev. A*, 52 (1995)
- SOME CONTROVERSY
- CW MODEL: Guoy effects
  - Y.Barad, et.al. *Appl. Phys. Lett.*, 70 (1997).
- DEBATE

  -interface sensitivity, or something else?
- FEMTOSECOND EFFECTS
  - D. Stoker, et.al. *Phys. Rev. A*, **71 061802** (2005).

<u>The origin of the interface sensitivity is group velocity mismatch of the ω/3ω pulses</u>.

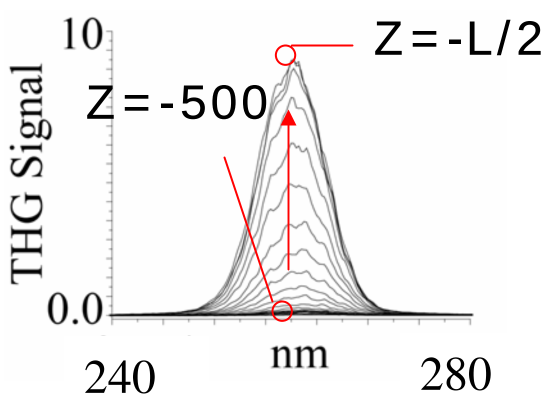


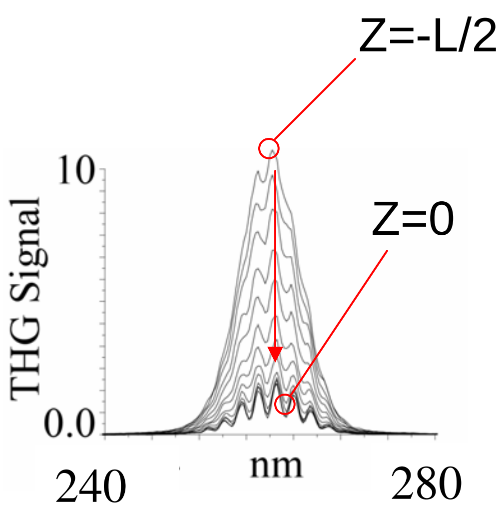


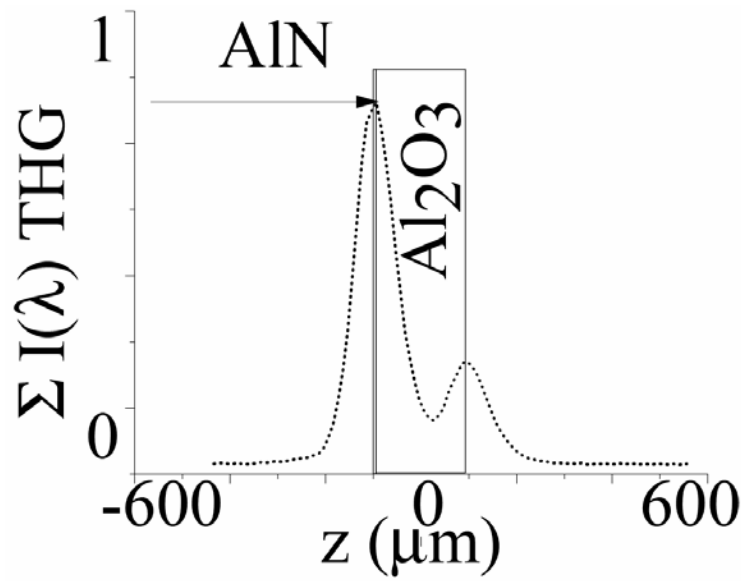
Spectrally resolving z-scans of thin films

provides clues about the nature of THG

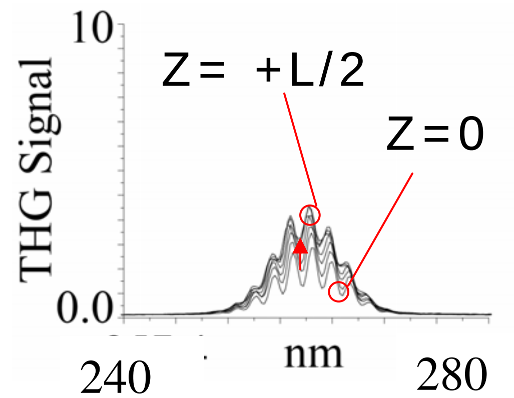
AlN coating

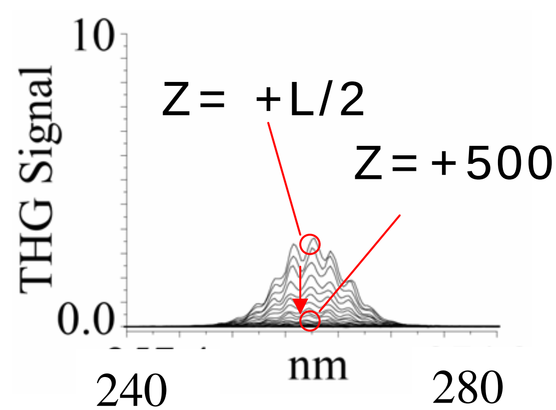






Uncoated substrate



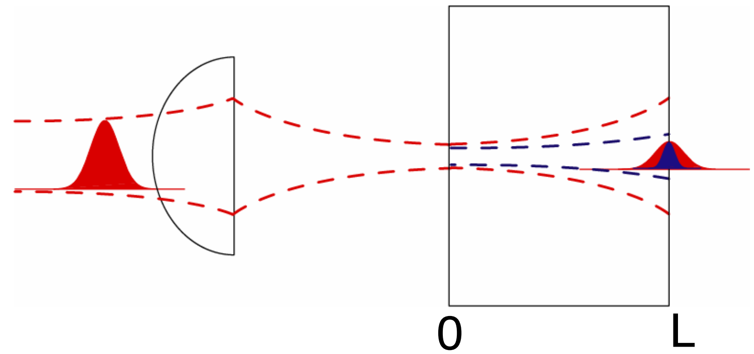


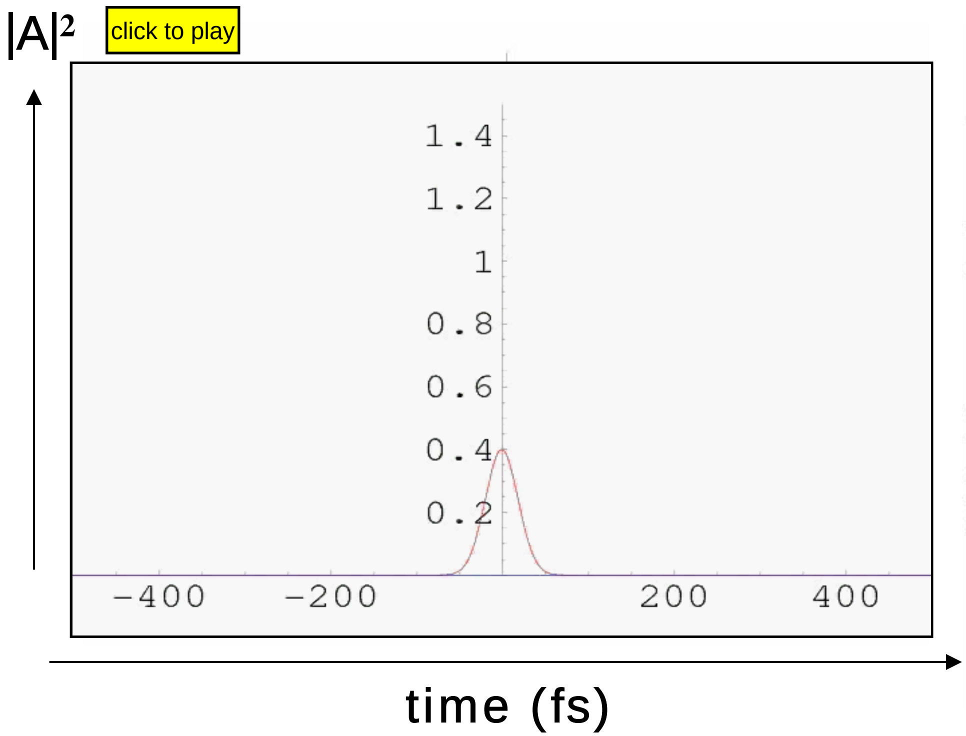
- Modulation Structure of Spectral Intensity comes only from the substrate
- Will show this is from GVM

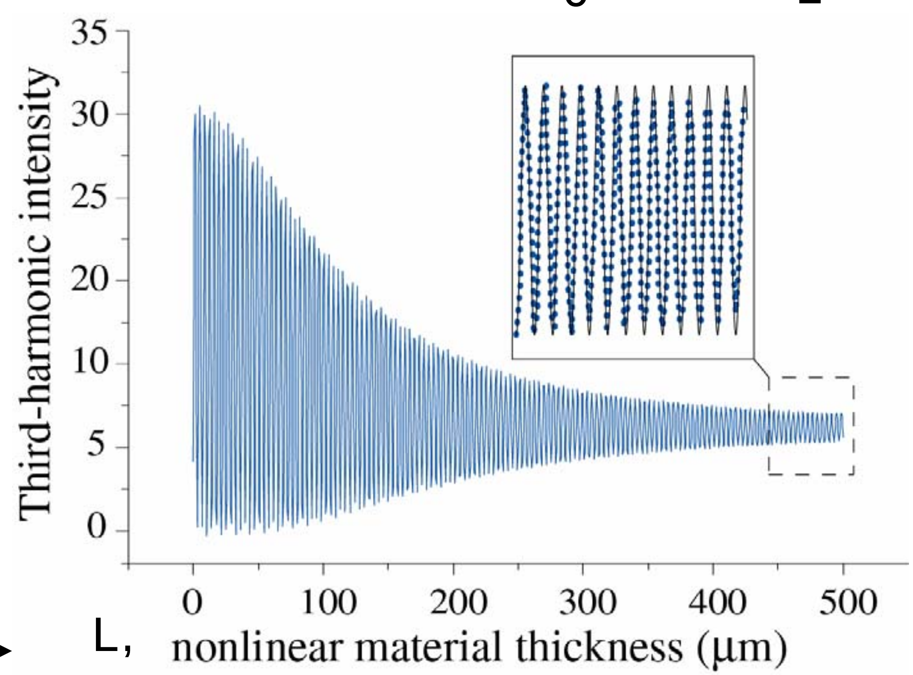
Numerical simulation of THG

for Gaussian focusing a continuous wave laser (BARAD's)-CW

$$\tilde{A}_q(z) \propto \int_0^L \frac{\chi^{(q)} A_0^q e^{i\Delta k z'}}{(1+i\frac{z'}{z_r})^{q-1}} dz'$$





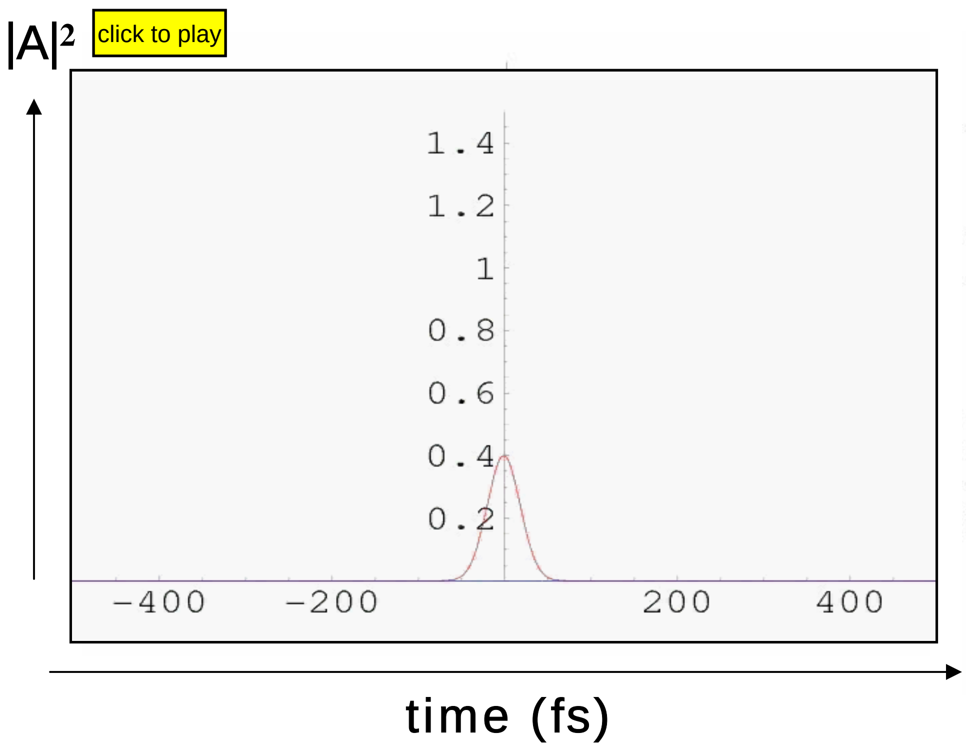


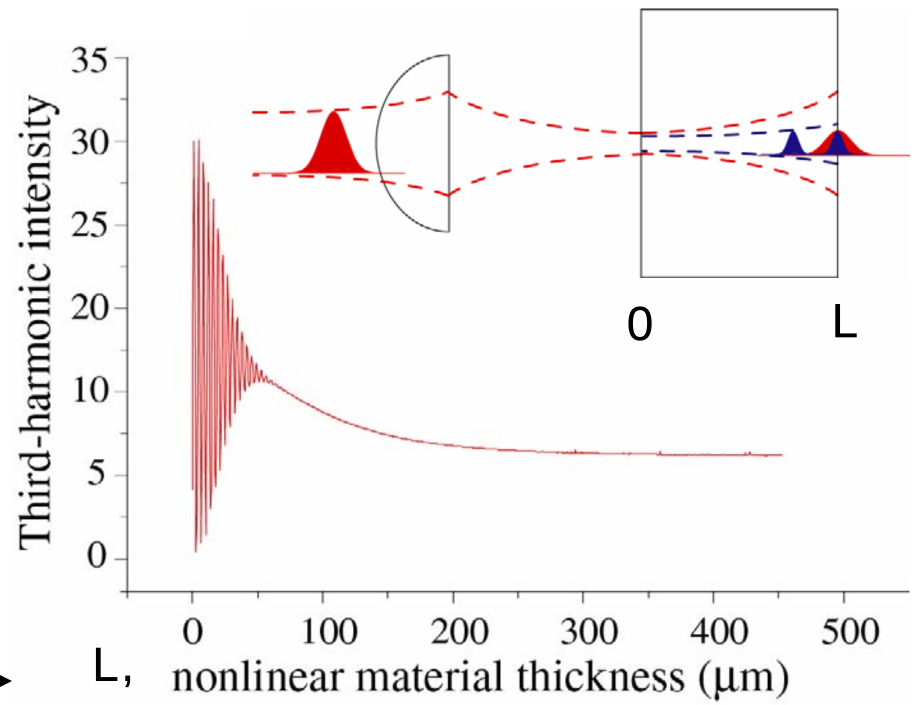
Numerical simulation of THG

for Gaussian focusing a femtosecond laser pulse, and including all time dependence due to GVM.

$\beta_1 = 1/v_g$

$$\tilde{A}_q(z,t') \propto \int_0^L \frac{\chi^{(q)}\left(A_0 e^{-\tilde{\Gamma}_0\left(t'-(\beta_1(3\omega_0)-\beta_1(\omega_0))z\right)^2}\right)^q e^{i\Delta k z}}{\left(1+i\frac{z}{z_r}\right)^{q-1}} dz$$





Calculating the THG spectrum

Is accomplished by relating the spectral intensity to the temporal intensity by a Fourier Transform

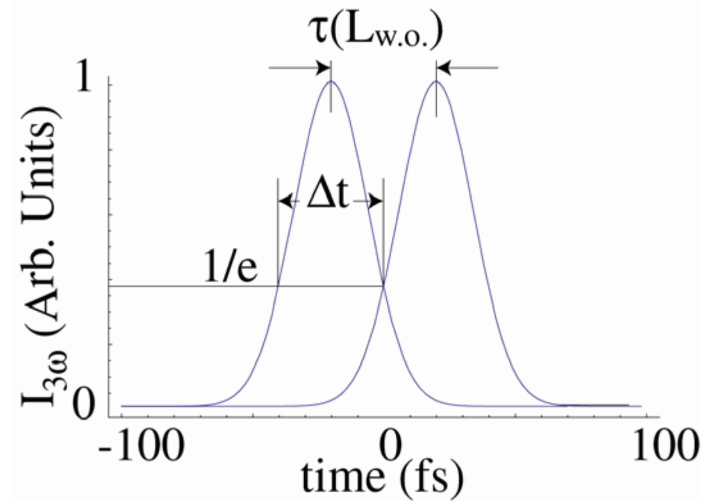
- Time domain: pulses are Gaussians, and separated in time by $\tau = (\beta_1(3\omega_0) - \beta_1(\omega_0))L$
- Field has amplitude proportional to intensity at each interface

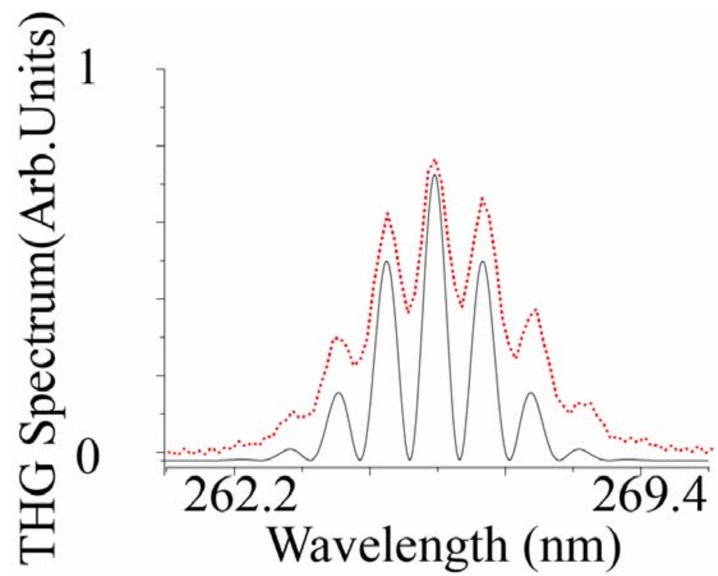
$$\tilde{A}(t) = e^{-\Gamma_0 t^2} + e^{-\Gamma_0 (t-\tau)^2}$$

- Fourier Transform of A:

$$|\tilde{A}(\Omega)|^2 = |\int_{-\infty}^{\infty} \tilde{A}(t) e^{-i\Omega t} dt|^2$$

$$|\tilde{A}(\Omega)|^2 = |\sqrt{\frac{\pi}{\Gamma_0}} e^{\frac{-\Omega^2}{4\Gamma_0}} (1 + e^{i\tau\Omega})|^2$$





Measuring a film's susceptibility

Is done by removing the substrate's contribution

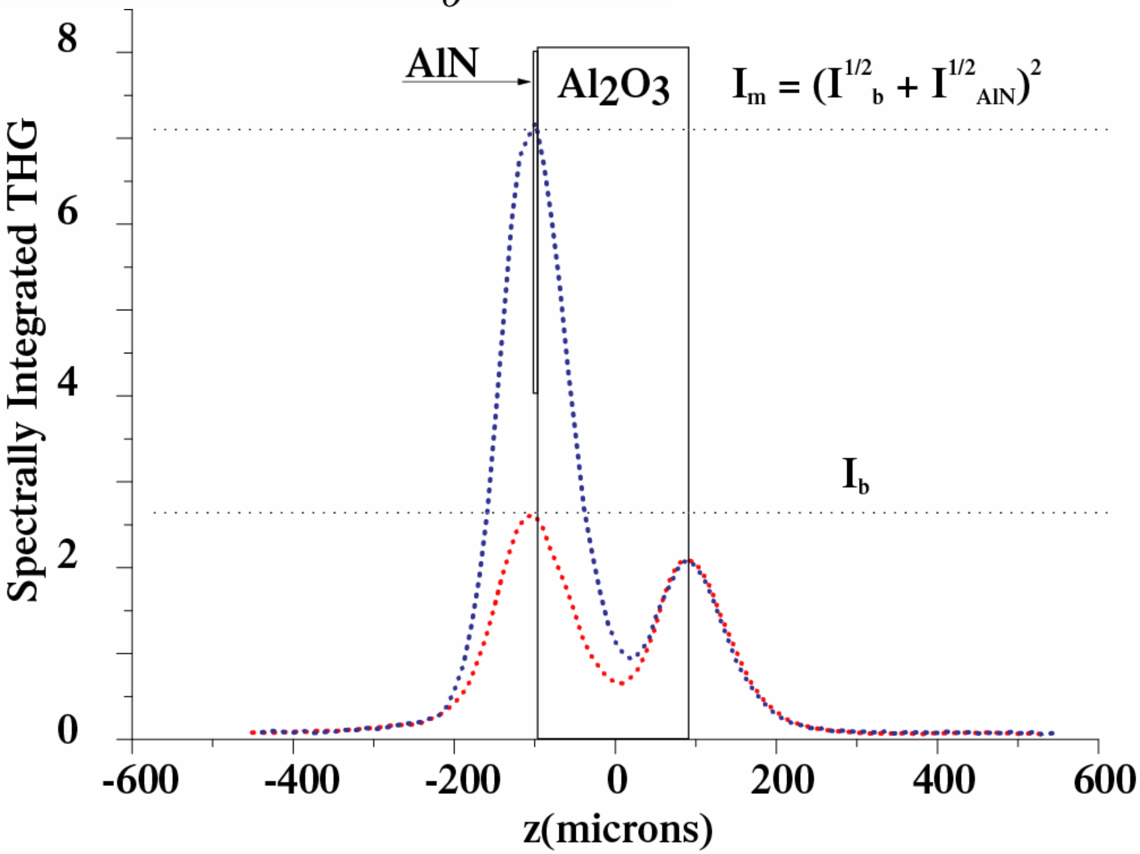
with the assumption, $E_{AlN+Al2O3} = E_{AlN} + E_{Al2O3}$, we can back-out the film's $\chi^{(3)}$

$$\chi^{(3)}_{AlN} = \chi^{(3)}_{Al_2O_3} \frac{J_p(Al_2O_3)}{J_p(AlN)} \left(\frac{I_m^{1/2}}{I_b^{1/2}} - 1\right)$$

"3 easy steps"

- 1) Measure bare substrate
- 2) Measure substrate with film
- 3) Use geometry

$$I_{3\omega} = \alpha |\chi^{(3)} J_p|^2 I^3_0$$

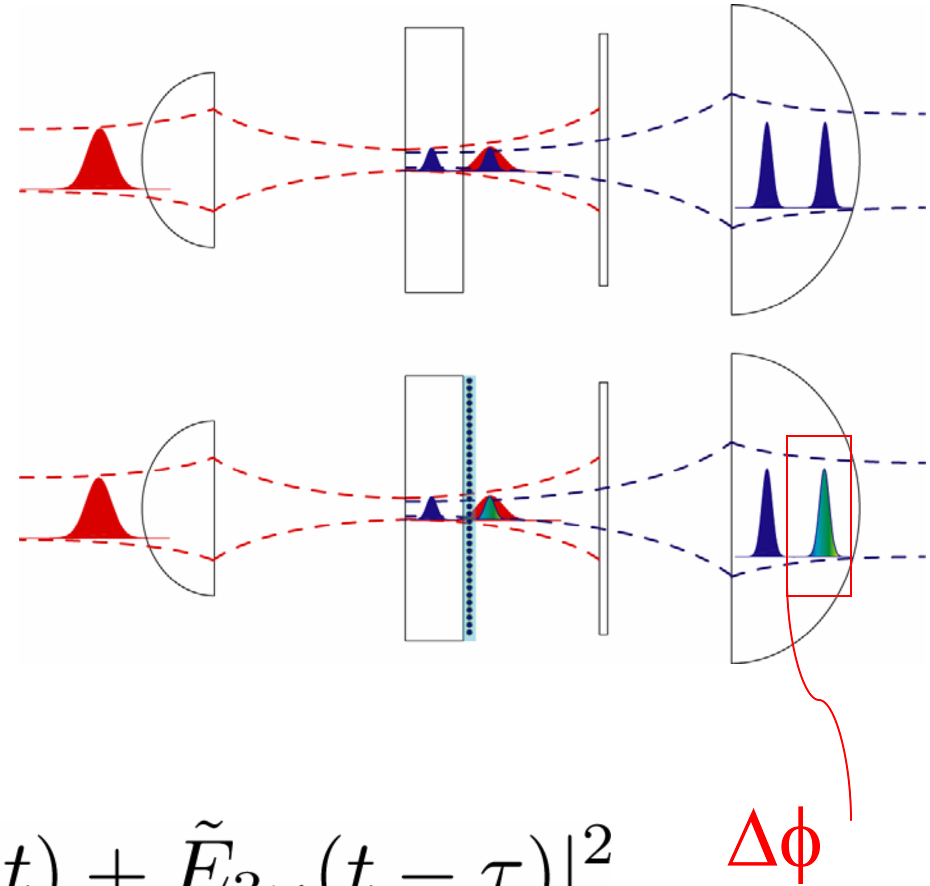


Absolute phase measurements

If the substrate thickness, L, satisfies $L \cong z_r$, consider the interferometric tool:

- Substrate is nonresonant, so there is no phase difference.

- Thin Resonant Film:
The interferometric pattern is altered when one interface contains a resonant material.

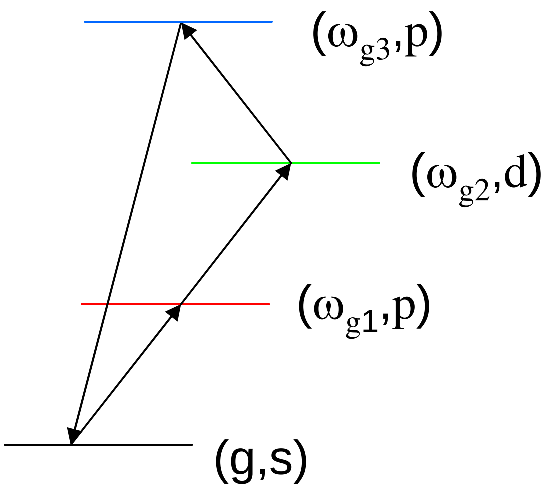


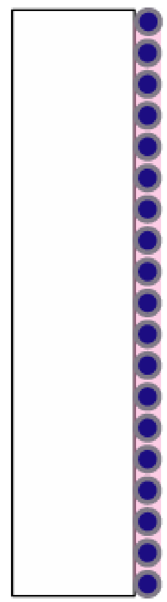
$$|I_{3\omega}(t)| \propto |e^{i\Delta\Phi}\tilde{E}_{3\omega}(t) + \tilde{E}_{3\omega}(t-\tau)|^2$$

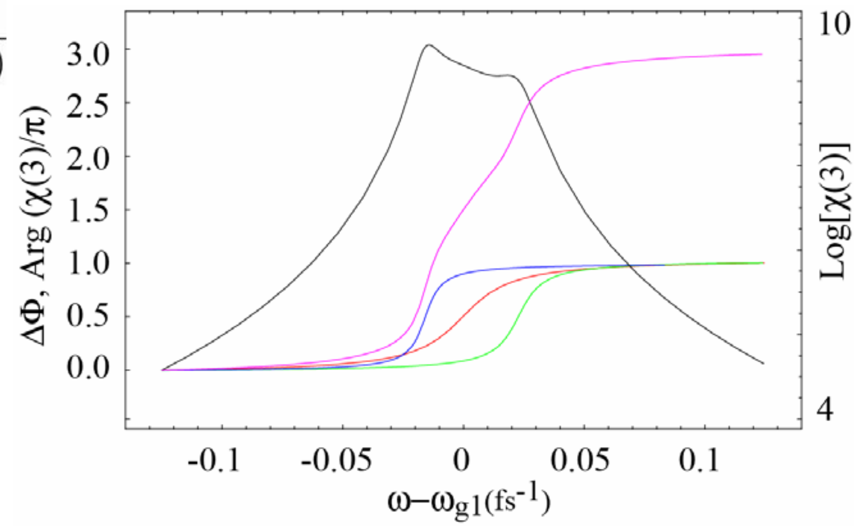
Interferometric Resonance-Generated Optical Third-Harmonic Spectroscopy

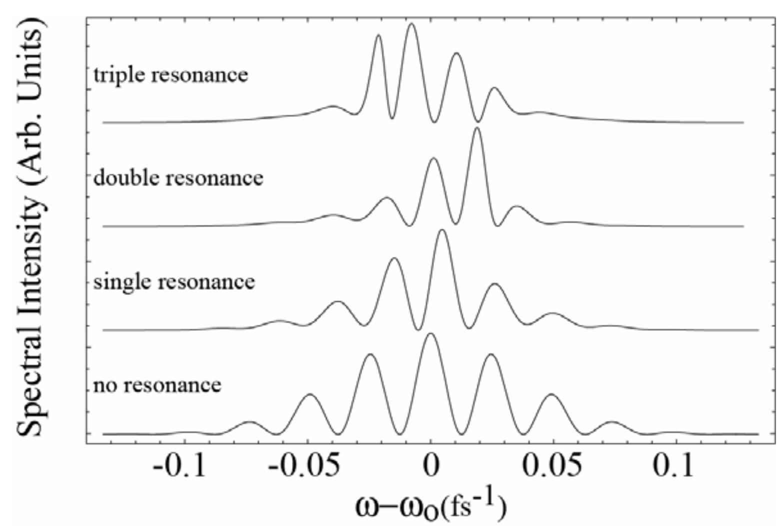
Are possible through measuring the spectral intensity <u>shifts</u> and can be used to characterize resonant materials

$$\chi^{(3)}_{res.} \propto \frac{1}{(\omega - \omega_{g1} + i\gamma_{g1})(2\omega - \omega_{g2} + i\gamma_{g2})(3\omega - \omega_{g3} + i\gamma_{g3})}$$









The electron carried through the closed n.l.o. cycle is *phase-shifted* by the argument of $\chi(3)$. The electron's phase is passed to the 3ω pulse.

Third-harmonic pulses are an ideal tool for studying multiphoton resonances in nanostructured optical materials

GROUP VELOCITY

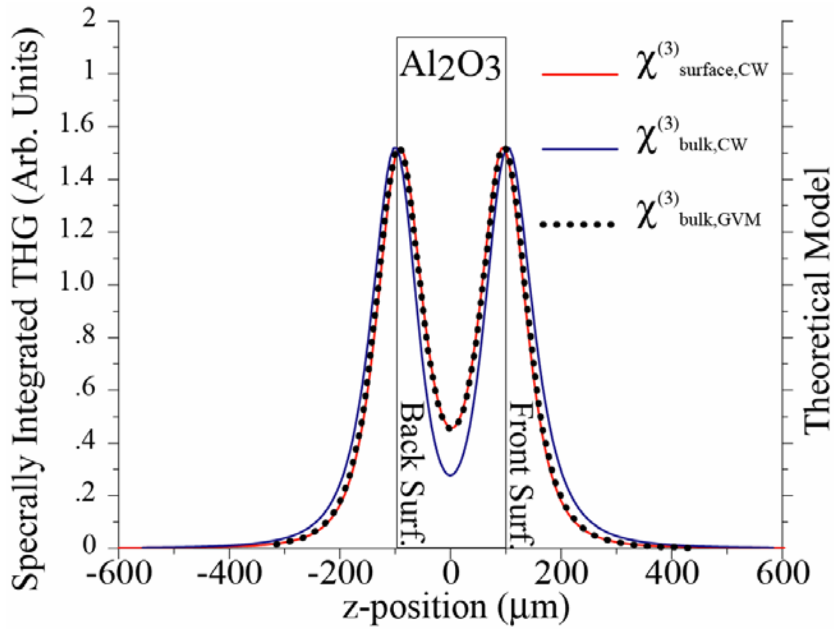
- effects can be very large

ZSCAN MEASUREMENTS

- appear to measure a surface susceptibility.

UNLIKE SURFACE SHG

- the signal does not originate from crystal asymmetry, but some changes in bulk parameters near an interface.



Using THG it is possible to measure the coherent transport (excitation, conduction) in nanostructured thin materials. We are currently pursuing applications for studying thin films made in our lab.

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