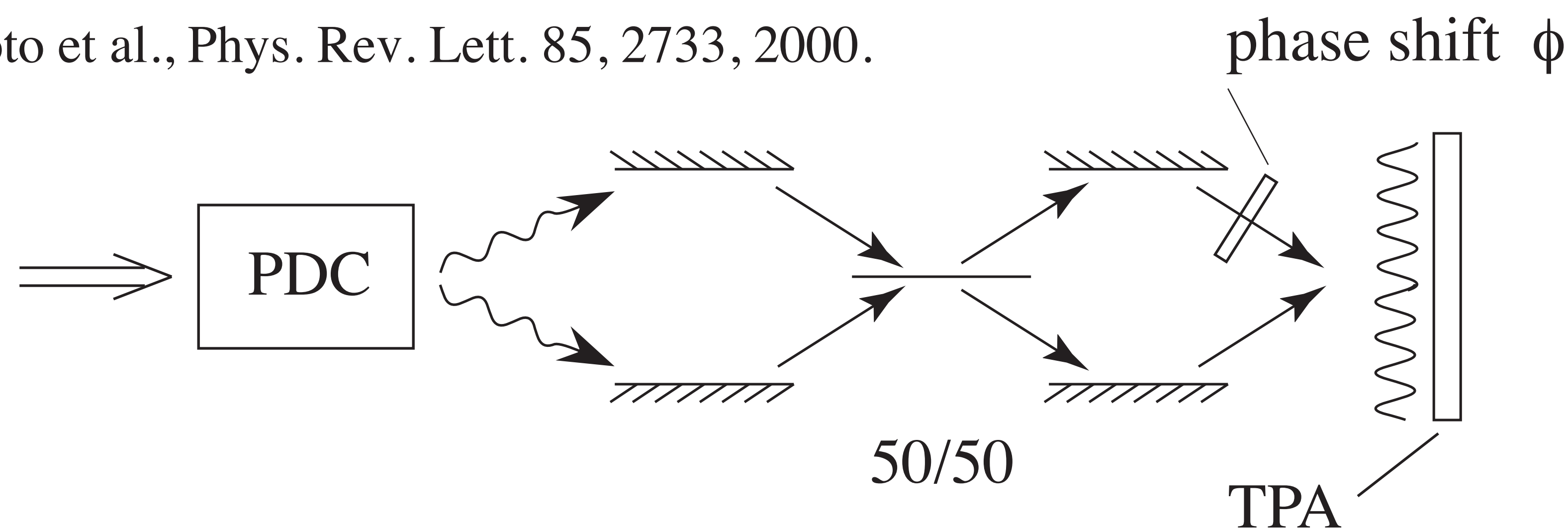
Quantum Lithography

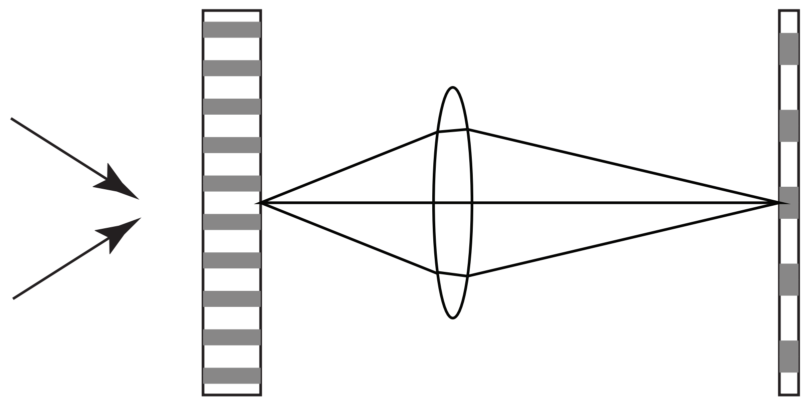
- Entangled photons can be used to form an interference pattern with detail finer than the Rayleigh limit
- Process “in reverse” performs sub-Rayleigh microscopy, etc.
- Resolution ≈ $\lambda / 2N$, where N = number of entangled photons

Boto et al., Phys. Rev. Lett. 85, 2733, 2000.



- No *compelling* laboratory demonstration to date
- Primary difficulty: need extremely sensitive recording material

Quantum “Lithography” – How to Observe?

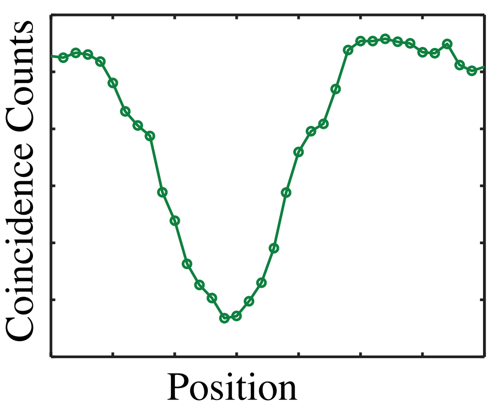


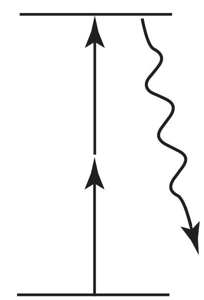
CCD camera

entangled photon pair

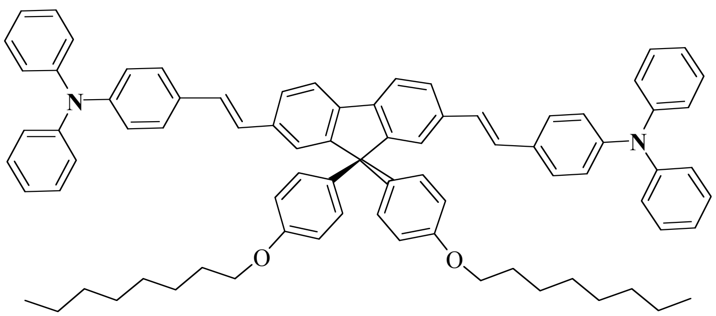
TPA followed by
short-wavelength fluorescence

HOM Dip





Two-Photon Absorbing Material



BOPF-TP

$C_{81}H_{80}N_2O_2$

4,4′-(1E,1′E)-2.2′-(9,9-bis(4-(octyloxy)phenyl)-9*H*-flourene-2,7-diyl)bis(ethene-2,1-diyl)bis(*N*,*N*-diphenylaniline

Excitation wavelength = 800 nm
Fluorescence wavelength = 459 nm

APPLIED PHYSICS LETTERS **89** (2006) 173133

Quantum Lithography – Materials Issues

What are the sensitivities of typical recording materials?

Silver halide holographic plates: 1 mJ/cm^2

Dichromated gelatin holographic plates: 100 mJ/cm^2

Two-photon photopolymer (Kawata): 1 MJ/cm^2

What typical values of multiphoton cross sections?

$\sigma^{(2)}$ typically 1 GM where 1 GM = 10^{-50} cm^2 s/photon

For a good molecular two-photon absorber, $\sigma^{(2)}$ = 1000 GM

For a SC QD two-photon absorber (Webb), $\sigma^{(2)}$ = 47,000 GM

We estimate that for PMMA $\sigma^{(3)} = 10^{-85}$ cm^4 s^2/photon

Can we do even better?

Good evidence that $\sigma^{(2)}$ and $\sigma^{(3)}$ can be enhanced by as much as 500-fold by coupling to a plasmonic resonance!

[Kano and Kawata, Opt. Lett, 21, 1848 1996;
Cohanoschi and Hernández, J. Phys. Chem. B 109, 14506 2005]

Enhanced Nonlinear Response through Microscopic Cascading

High-order NLO effects are typically much weaker than low-order effects

We can sythesize high-order response from repeated low-order response

This procedure is known as cascading, e.g., $\chi^{(3)}_{\mathrm{eff}} = \mathrm{const} \times \chi^{(2)} : \chi^{(2)}$

Cascading can be either macroscopic, which involves propagation effects

Example ω + ω creates 2ω; then 2ω + ω creates 3ω

Or it can be microscopic: two adjacent atoms can interact by means of "local field effects" to create a high-order response.

We have recently predicted a new consequence of local field effects which could lead to efficient high-order NLO processes, and now have data demonstrating this effect. [Dolgaleva, Boyd, Sipe, PRA 76, 063806 (2007)]

We hope to create efficient three-photon absorbers out of two-photon absorbers! Recall $\sigma^{(2)}$ is proportional to $\chi^{(3)}$; $\sigma^{(3)}$ proportional to $\chi^{(5)}$.

Experimental Separation of Microscopic Cascading Induced by Local-Field Effects

Ksenia Dolgaleva, Heedeuk Shin, and Robert W. Boyd

Institute of Optics, University of Rochester

John E. Sipe

Department of Physics, University of Toronto

FiO 2008 Tuesday, October 21, 11:15 a. m.

Lorentz Local Field

$$\boldsymbol{E}_{\text{loc}} = \boldsymbol{E} + \frac{4\pi}{3}\boldsymbol{P}$$

or

$$\boldsymbol{E}_{\text{loc}} = L\,\boldsymbol{E}$$

where

$$L = \frac{\epsilon^{(1)} + 2}{3}$$

is Lorentz local-field correction factor

$\epsilon^{(1)}$ - dielectric permittivity

J. D. Jackson, "Classical Electrodynamics"

Local Field in Nonlinear Optics

Local-field-corrected

$$P = \chi E = \chi^{(1)} E + 3\chi^{(3)} |E|^2 E + 10\chi^{(5)} |E|^4 E + \ldots$$

Local Field in Nonlinear Optics

$$\chi^{(1)} = N\,\gamma_{\text{at}}^{(1)}\,L;$$

$$\chi^{(3)} = N\,\gamma_{\text{at}}^{(3)}\,|L|^2\,L^2.$$

$\gamma_{\text{at}}^{(i)}$ - *i*-th nonlinear microscopic hyperpolarizability

Local Field in Nonlinear Optics

Since

$$\chi^{(3)} = N\,\gamma_{\mathrm{at}}^{(3)} |L|^2 L^2,$$

one would think that

$$\chi^{(5)} = N\,\gamma_{\mathrm{at}}^{(5)} |L|^4 L^2.$$

Microscopic Cascading by Local-Field Effects

"direct" contribution from fifth-order hyperpolarizability $\gamma_{\text{at}}^{(5)}$

$$\chi^{(5)} = N\gamma_{\text{at}}^{(5)}|L|^4 L^2 + \frac{24\pi}{10} N^2 (\gamma_{\text{at}}^{(3)})^2 |L|^4 L^3 + \frac{12\pi}{10} N^2 |\gamma_{\text{at}}^{(3)}|^2 |L|^6 L .$$

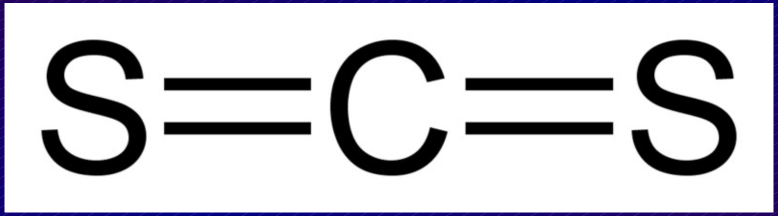
"microscopic cascaded" contributions from third-order hyperpolarizability $\gamma_{\text{at}}^{(3)}$

K. Dolgaleva, R. W. Boyd, J. E. Sipe, Phys. Rev. A **76,** 063806 (2007).

Experiment on Separation of Microscopic Cascaded Contribution

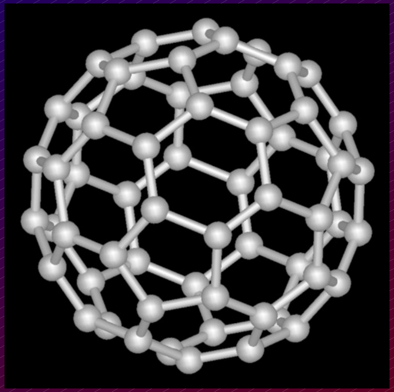
Changing the concentration of fullerene C_{60} in CS_2, we measured $\chi^{(5)}$ as a function of N.

Carbon disulfide

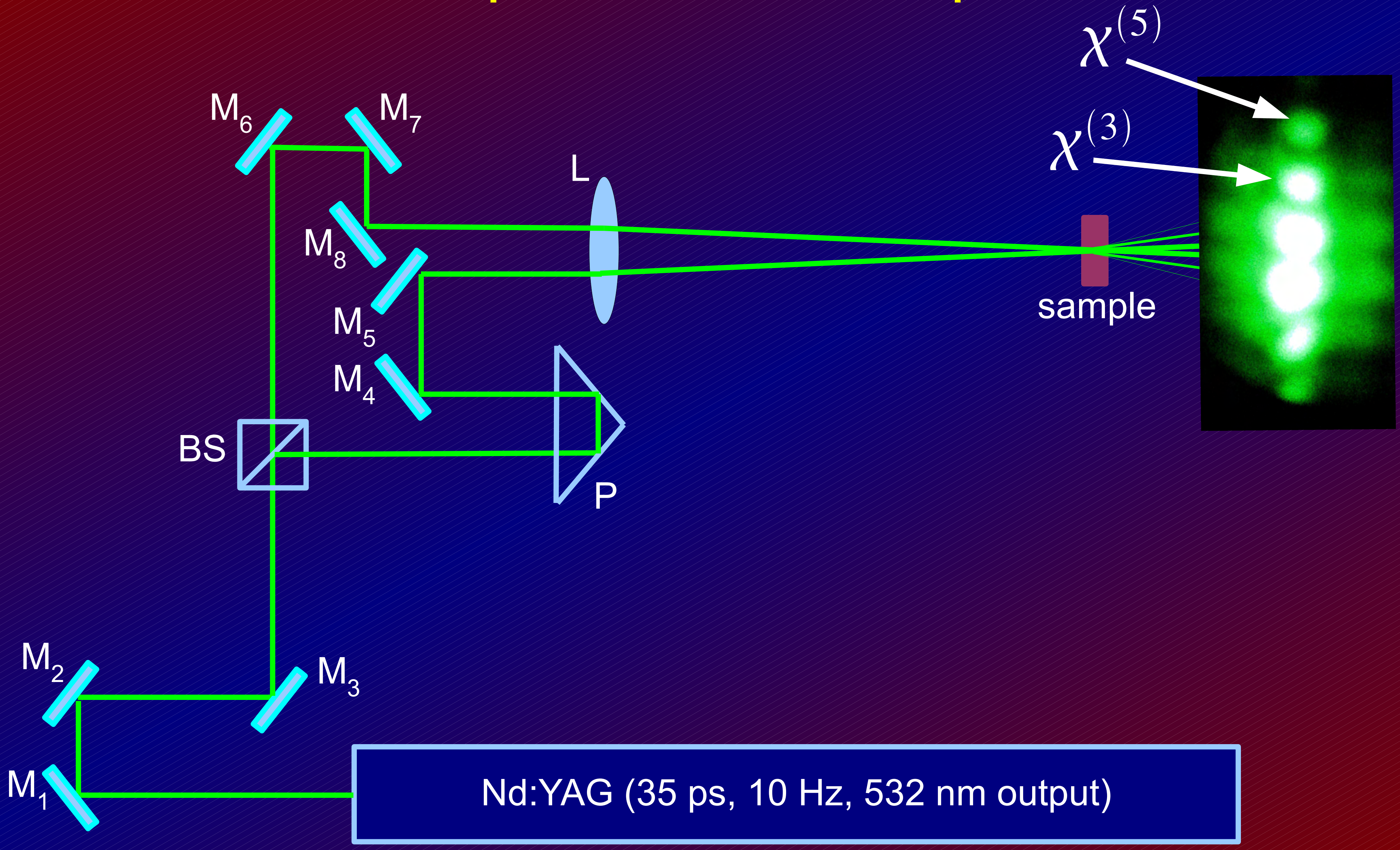


+

Fullerene C_{60}



Experimental Setup



Conclusions

- There is a microscopic cascaded contribution to $\chi^{(5)}$ induced purely by local-field effects.
- We performed an experiment to identify the microscopic cascaded contribution to $\chi^{(5)}$ and found that, under certain conditions, the value of this contribution can be larger than that of the macroscopic cascaded term.
- Microscopic cascading can induce high-order nonlinearities useful for quantum information.

Coherence and Indistinguishability in Two-Photon Interference

Anand Kumar Jha, Malcolm N. O'Sullivan-Hale,
Kam Wai Chan, and Robert W. Boyd

Institute of Optics, University of Rochester

What are the relevant degrees of freedom of a biphoton?

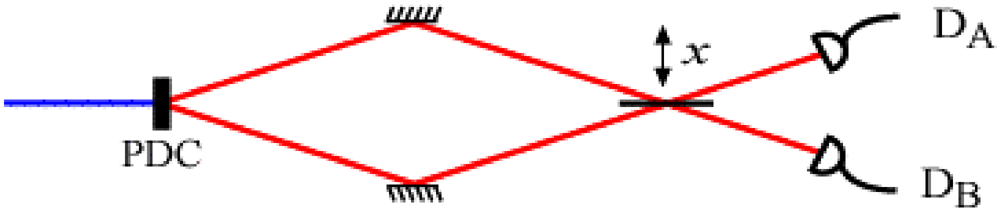
What are the generic features of two-photon interference?

Phys. Rev. A, 77 021801 (R) (2008)

Two-Photon Interference -- How to Understand?

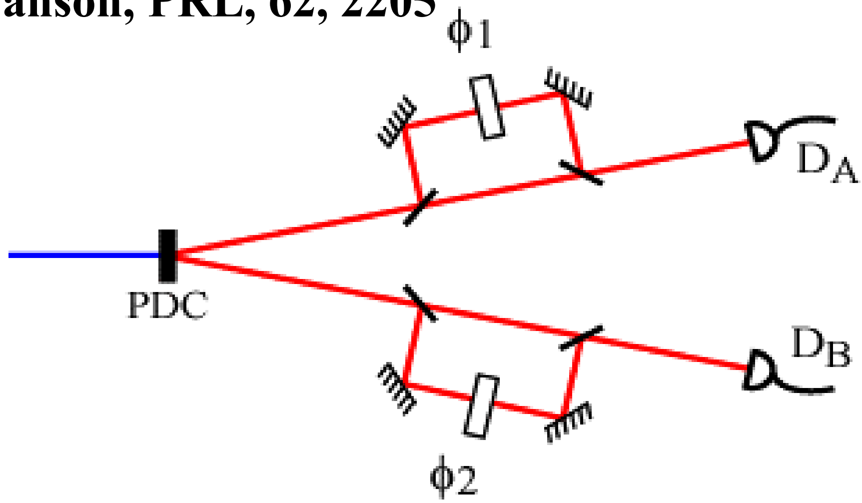
- **Hong-Ou-Mandel effect (1987)**

PRL, 59, 2044



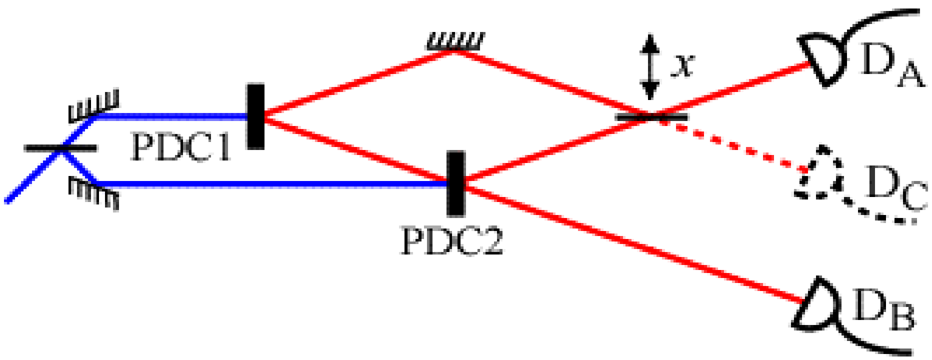
- **Bell Inequality for position and time (1989)**

Franson, PRL, 62, 2205



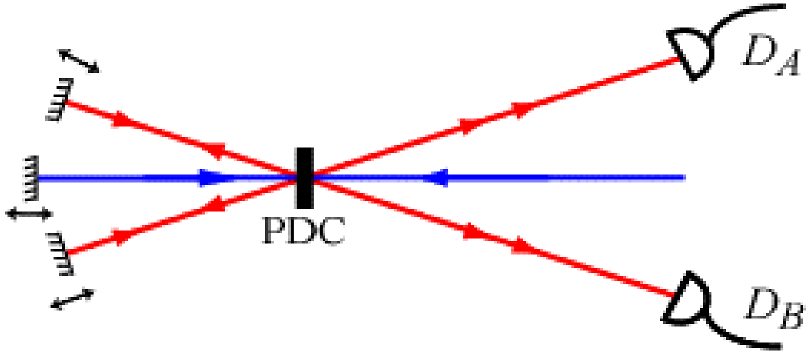
- **Induced Coherence (1991)**

Zou et al. PRL, 67, 318

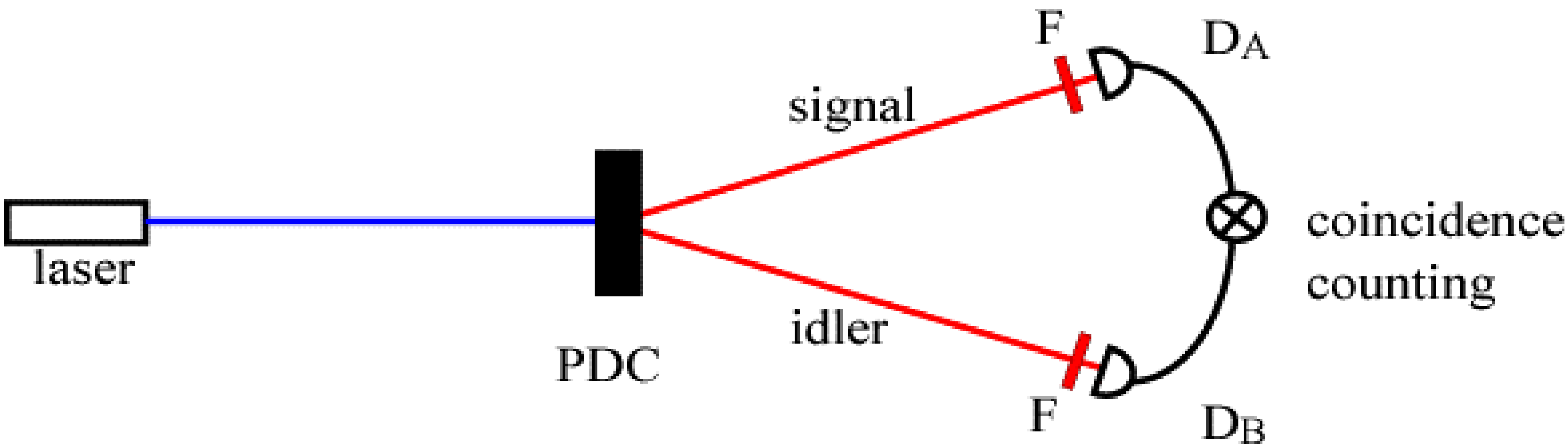


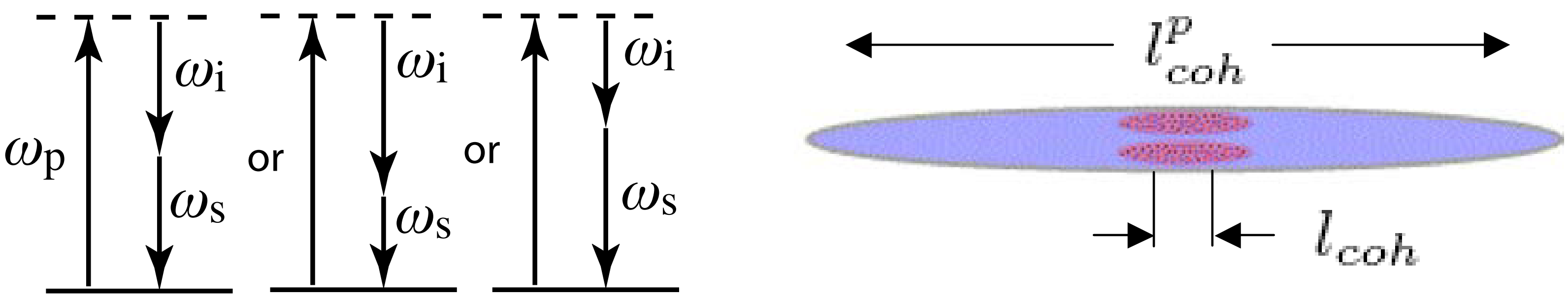
- **Frustrated two-photon creation (1994)**

Herzog et al. PRL, 72, 629



Biphotons Are Created by Parametric Downconversion (PDC)



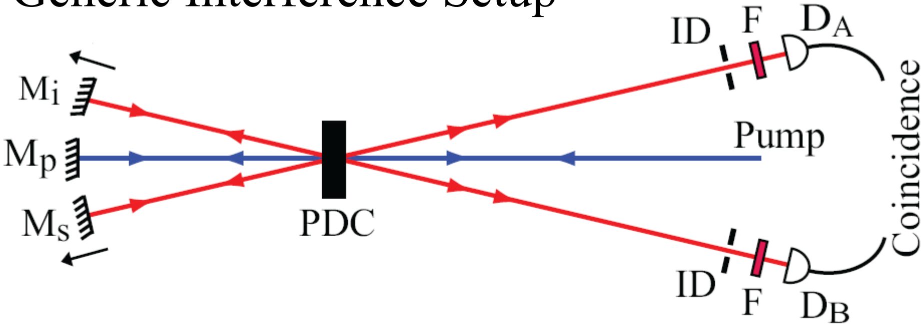


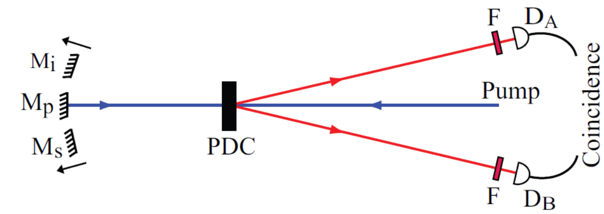
Length of two-photon wavepacket ~ coherence length of pump laser ~ 10 cm

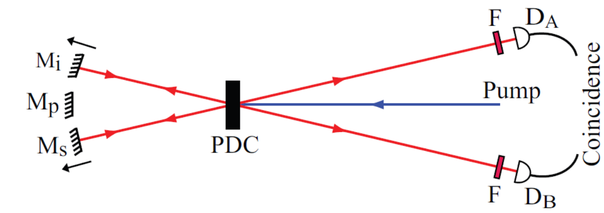
Coherence length of signal/idler photons ~ c/Δω ~ 100 μm.

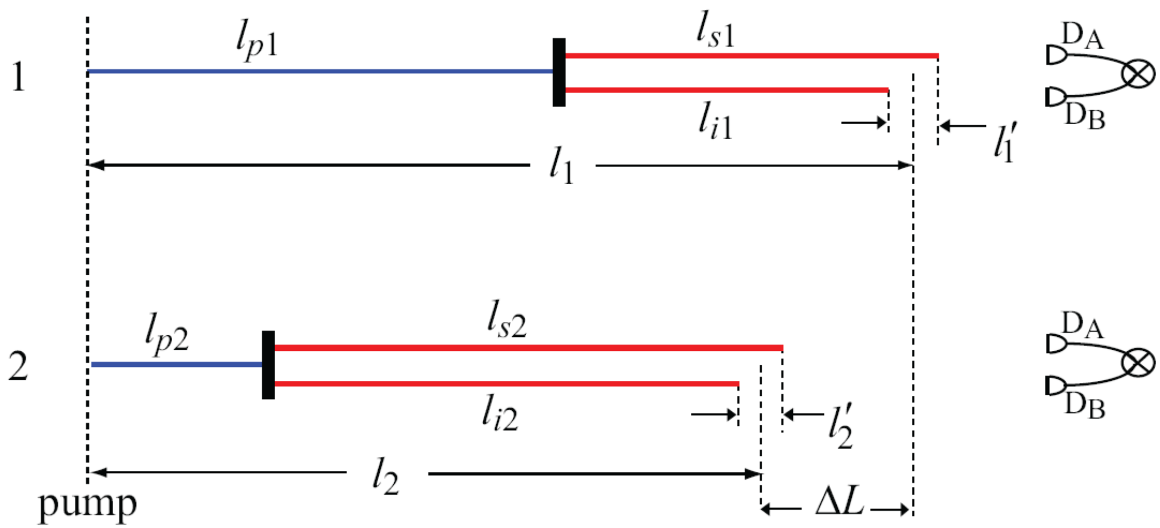
What Are Coherence Requirements for Two-Photon Interference ?

Generic Interference Setup









$$\Delta L \equiv l_1 - l_2$$

Biphoton path-length

$$\Delta L' \equiv l_1' - l_2'$$

Biphoton path-asymmetry length

$$R_{AB} = C\left[1 + \gamma'\left(\Delta L'\right)\gamma\left(\Delta L\right)\cos\left(k_0 \Delta L\right)\right]$$

Jha et al., PRA 77, 021801(R) (2008)

Necessary conditions for two-photon interference:

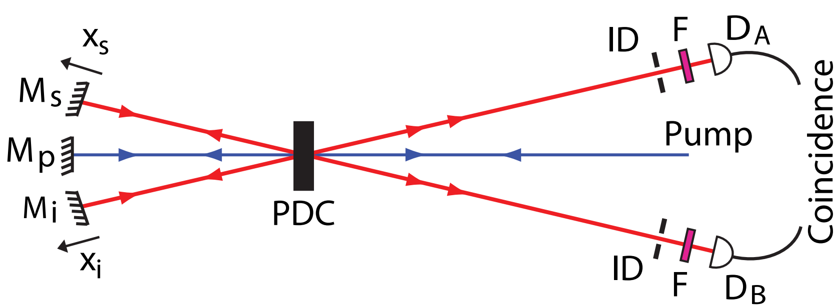
$$\Delta L < l^p_{\text{coh}}$$

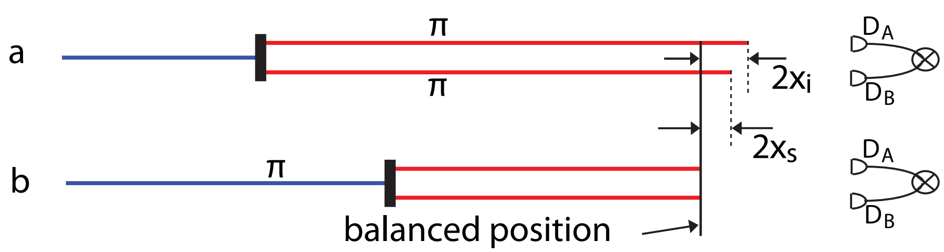
$$\Delta L' < l_{\text{coh}}$$

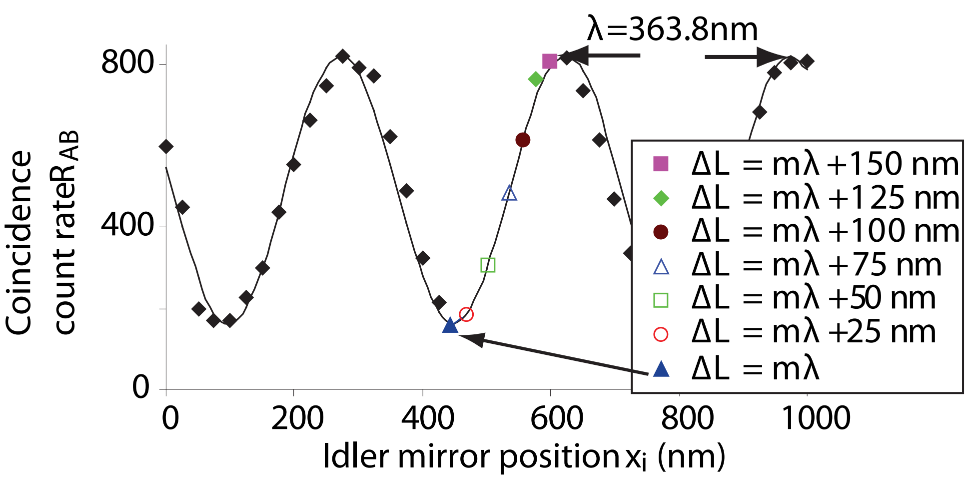
$l^p_{\text{coh}} \sim 10$ cm

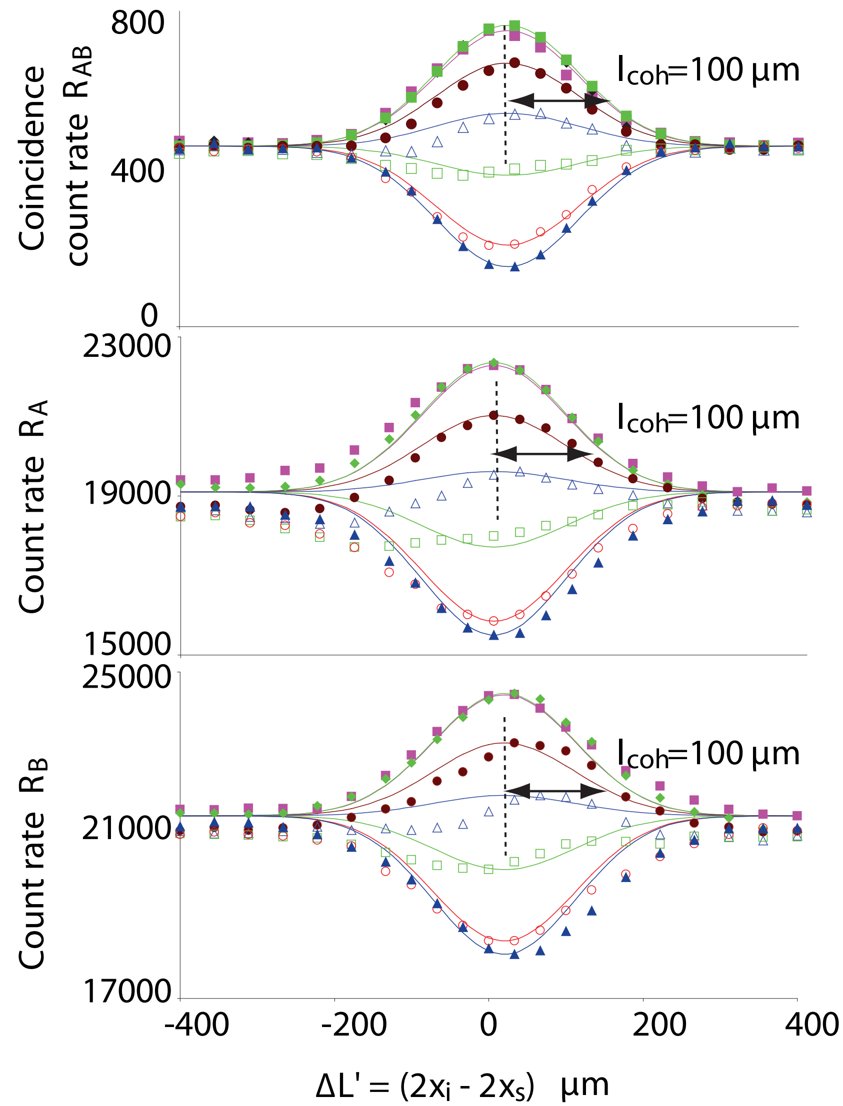
$l_{\text{coh}} = \dfrac{c}{\Delta\omega} \sim 100\ \mu\text{m}$

Our Experiment: Generalization of the Hong-Ou-Mandel Effect







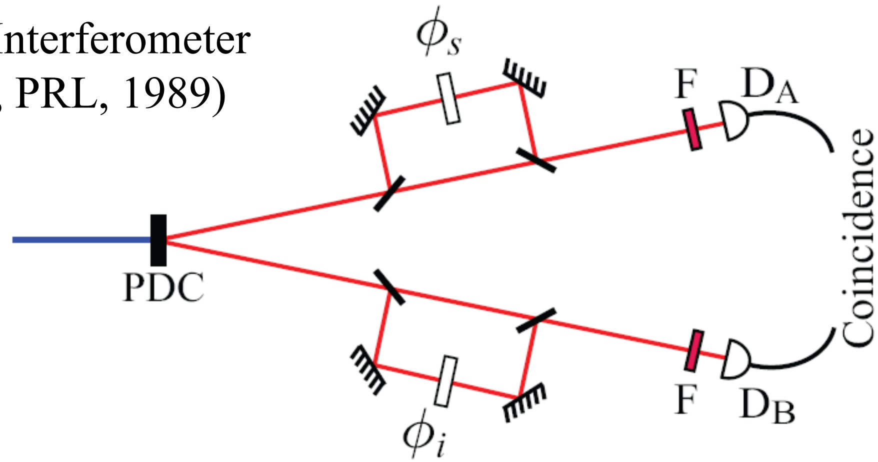


Jha et al., PRA 77, 021801(R) (2008).

We see either a dip or a hump (depending on the value of ΔL) in both the single and coincidence count rates as we scan $\Delta L'$.

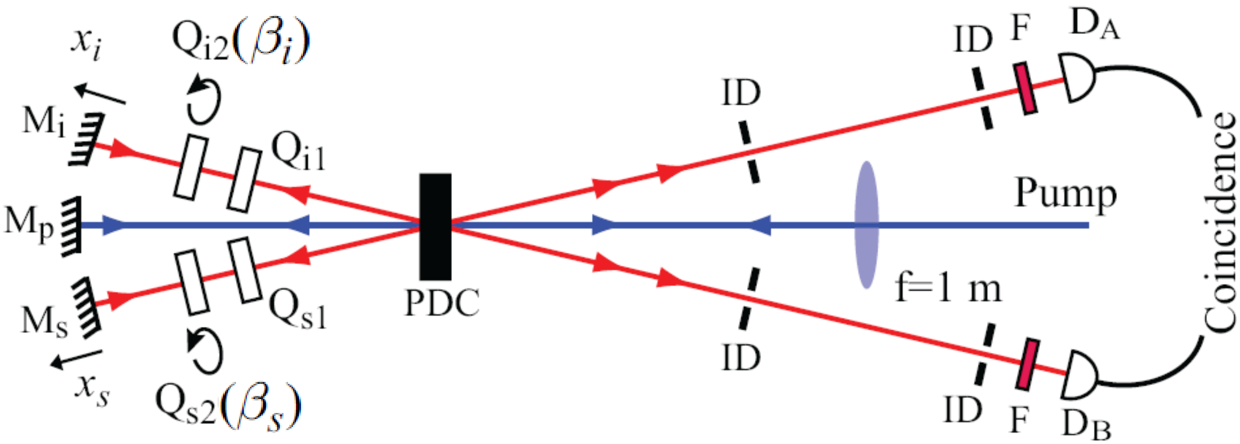
Bell Inequality for Energy-Time Entanglement Controlled by Geometric (Berry's) Phase

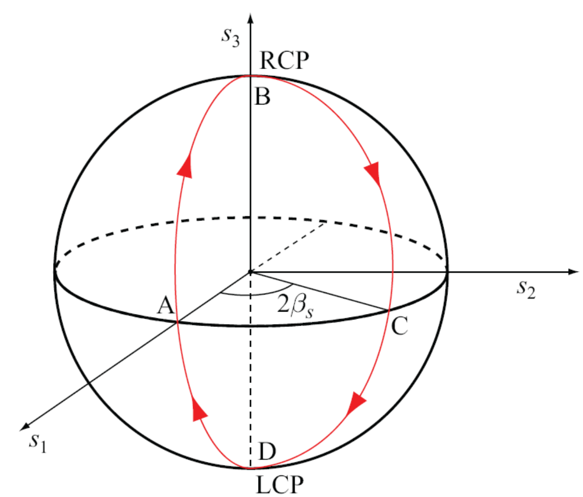
Franson Interferometer
(Franson, PRL, 1989)



$$R_{AB} = C[1 + \cos(\phi_s + \phi_i)]$$

Violation of CHSH Bell Inequality using dynamic phase





$$R_{AB} = C\ \{\ 1 - \cos[k_0(x_s + x_i) + 2\beta_s + 2\beta_i]\}$$

Violation of CHSH Bell Inequality using geometric (Pancharatnam, Berry) phase

Jha, Malik, Boyd, PRL 101, 180405 (2008).