A Proposal to use Intensity Interferometry / Multiphoton Correlation Measurements to Solve the Phase Problem in X-ray Crystallography

Or: light is weirder than you think

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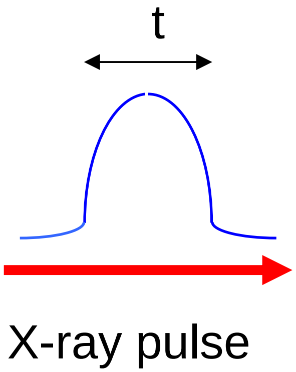
LBL
In collaboration with M. Howells, J. Holton

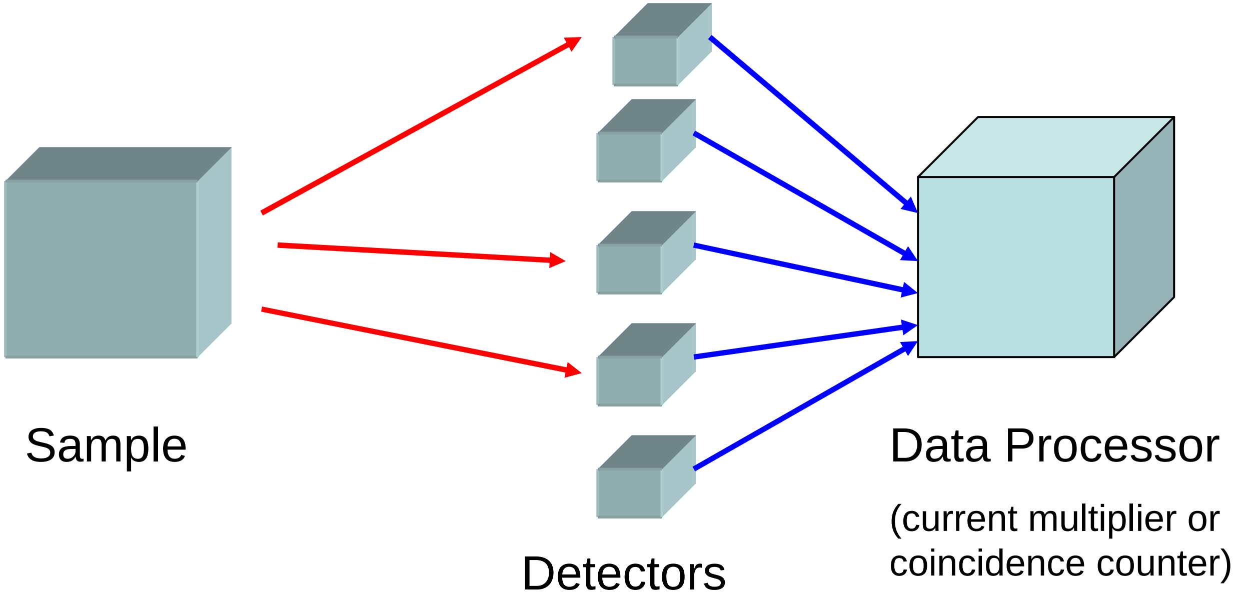
Supported by DOE IDAT grant

Why?

- Phase information can theoretically be obtained by measuring 2 or more “simultaneous” photons in coincidence
- This is all preliminary work! (after >2.5 years)


t

X-ray pulse


Sample

Detectors

Data Processor

(current multiplier or coincidence counter)

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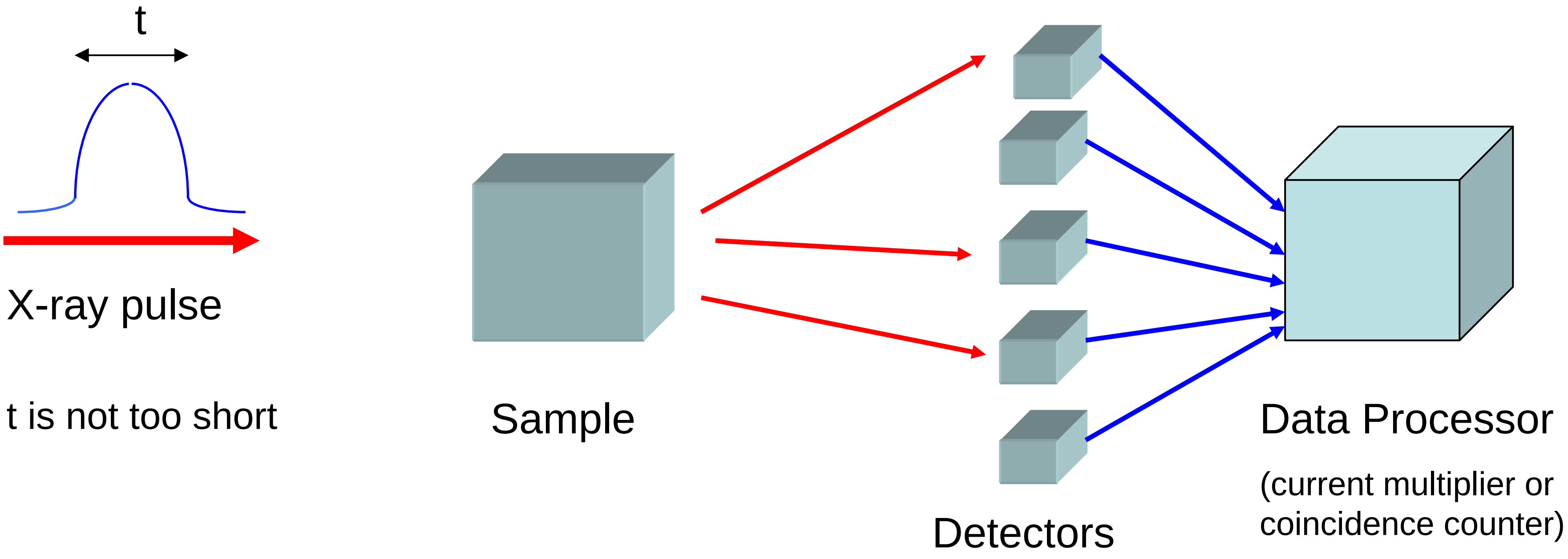

t

X-ray pulse

t is not too short

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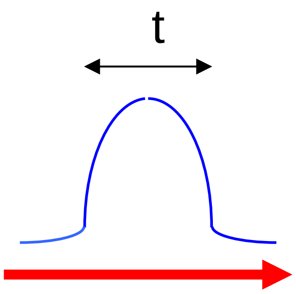
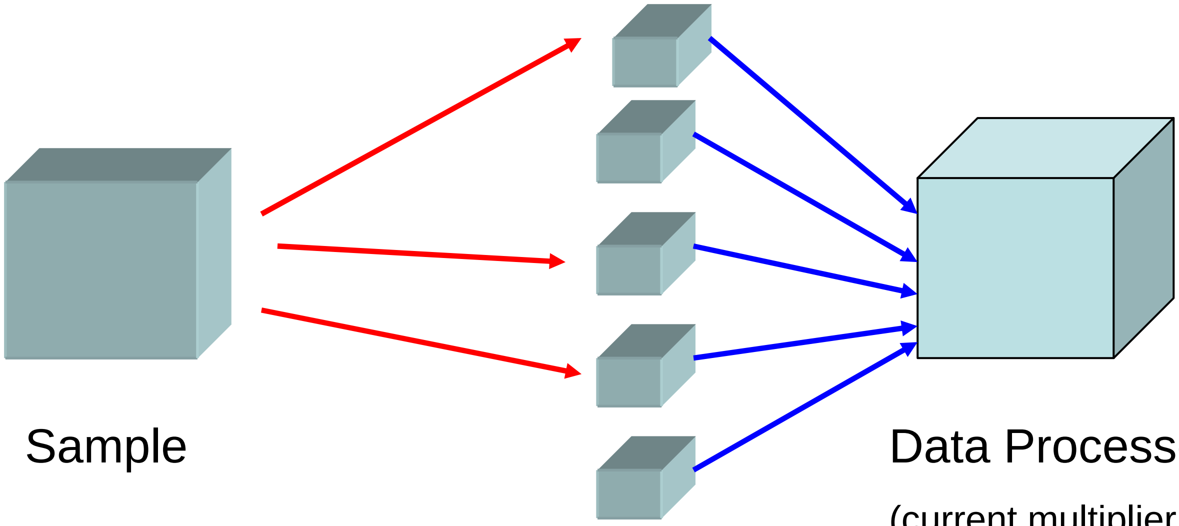


X-ray pulse

t is not too short

not too long



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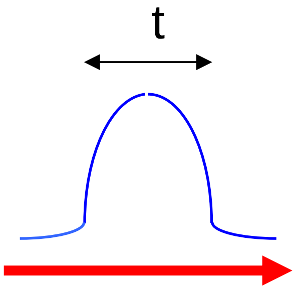
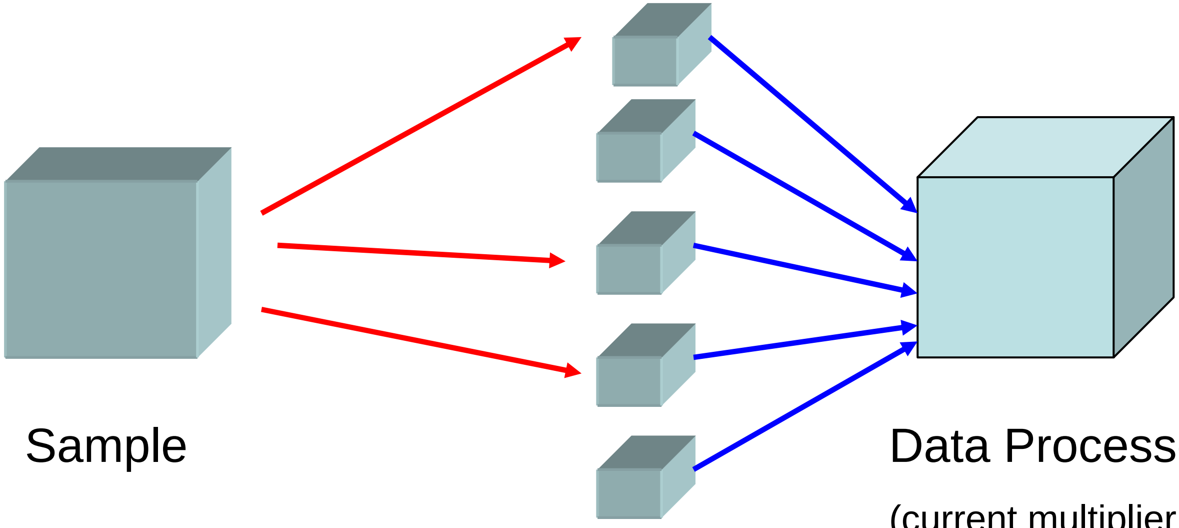


X-ray pulse

t is not too short

not too long

but just right



Sample

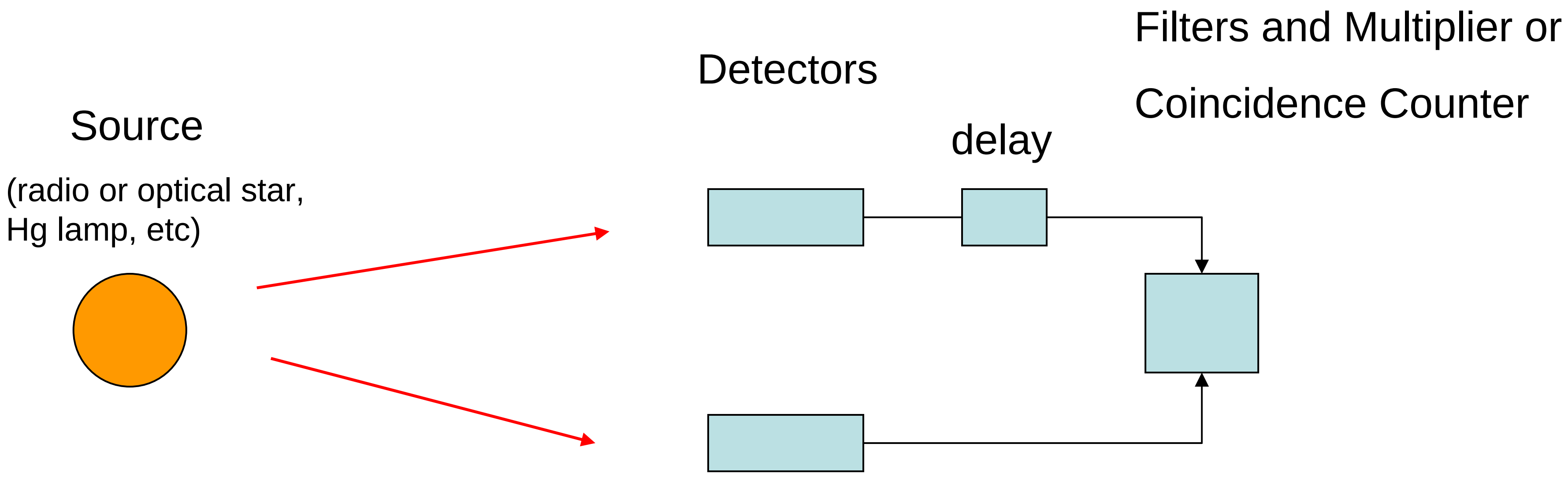
Detectors

Data Processor

(current multiplier or coincidence counter)

Intensity Interferometer/Photon Correlations

Hanbury-Brown Twiss Effect



Filters and Multiplier or

Coincidence Counter

Detectors

delay

Source

(radio or optical star,
Hg lamp, etc)

- Incoherent source – no lasers, please
- Detectors: Radio Receiver, Photomultiplier,PD, APD, etc. + filters
- A radio receiver can be described classically, a photomultiplier and other devices must take detector response into account
- Hg lamp experiment uses a beam splitter so intensities are identical at each detector

Background

- Can intensity interferometry/multiphoton correlation measurements be used to solve the phase problem in protein crystallography (or single molecule measurements?)
- Early responses by the “experts”:
  1) “Been there, done that”; (HENPs, GLW)
  2) “Photon antibunching and entanglement are more interesting (quantum optics)
  3) “It’s obvious, go do the experiment”;
  4) “That’s an important problem”;
  5) “It might work, let me think about it”
- Review of the literature suggests idea may not even be novel, but getting the experiment to work might be novel!
- Work continues on the feasibility
- Apologies in advance to those we have misinterpreted
- Almost everything I present was/is <u>controversial</u>!
- And some may be wrong!

Outline

- Description of an Intensity Interferometer
- Optical Coherence and a little theory
- Scattering Theory approach
- How to get phase information
- High energy, nuclear physics experiments
- Accelerator experiments – real experiments that have been done!
- Imaging experiments and theory (Optical)
- Design ideas for crystallography and test experiments

- Imaging, scattering, diffraction different ways of looking at the same physics
- Equations minimized – see http://bl1231.lbl.gov/~sibyls/Pickup
- See me for a list of the some of the most relevant papers

Intensity Interferometer

- Hanbury-Brown and Twiss built an intensity interferometer to measure the angular size of stars (radio and visible frequencies)
- The technique utilizes measurement of intensity fluctuations
  "using noise to measure the size of stars"
- They also did lab experiments with a Hg lamp source
- Lab experiments need a bright, narrow band source; otherwise it can take 10^3 to 10^{11} years to get a signal – those Nature papers are too short, often just results, few details!
- Incoherent source, individual measurements fast and light monochromatic enough so that the light is partially coherent – confounding and giving nightmares (and opportunities) to theorists and experimentalists alike
- Physics nonintuitive or counterintuitive (for a Hg lamp, a coincidence experiment must use less bright light than a current multiplier experiment – due to time resolution of electronics?)
- Describable by classical theory (+ photoelectron detector if used; need for Scully-Lamb or Ueda photodetector theory and master equations?) or quantum theory

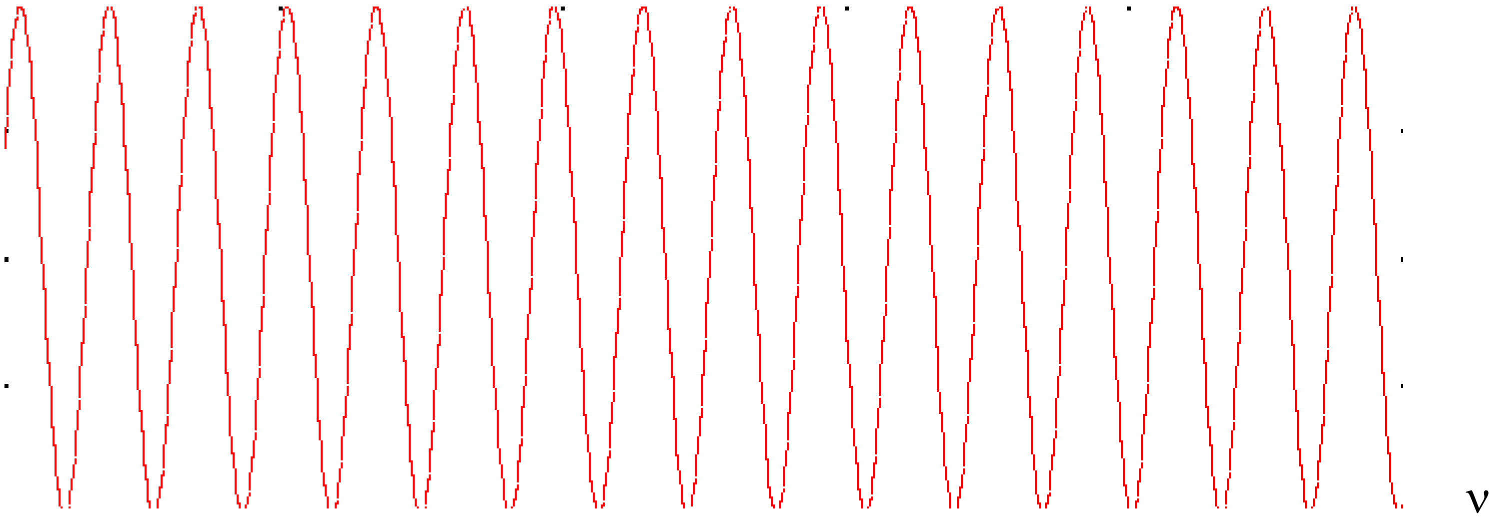
Theory (simplified)

- It all started with Einstein (with Hopf (1910) and Von Laue (1915, 1907)
- White light of thermal origin has the properties of a Gaussian random process – which makes the theorists happy (that makes for a whole lot of independently generated photons!)
- Then the electric field can by represented by a complex analytic field V(t) and the time average intensity is

  $$\frac{1}{2} \langle V^*(t)V(t) \rangle$$

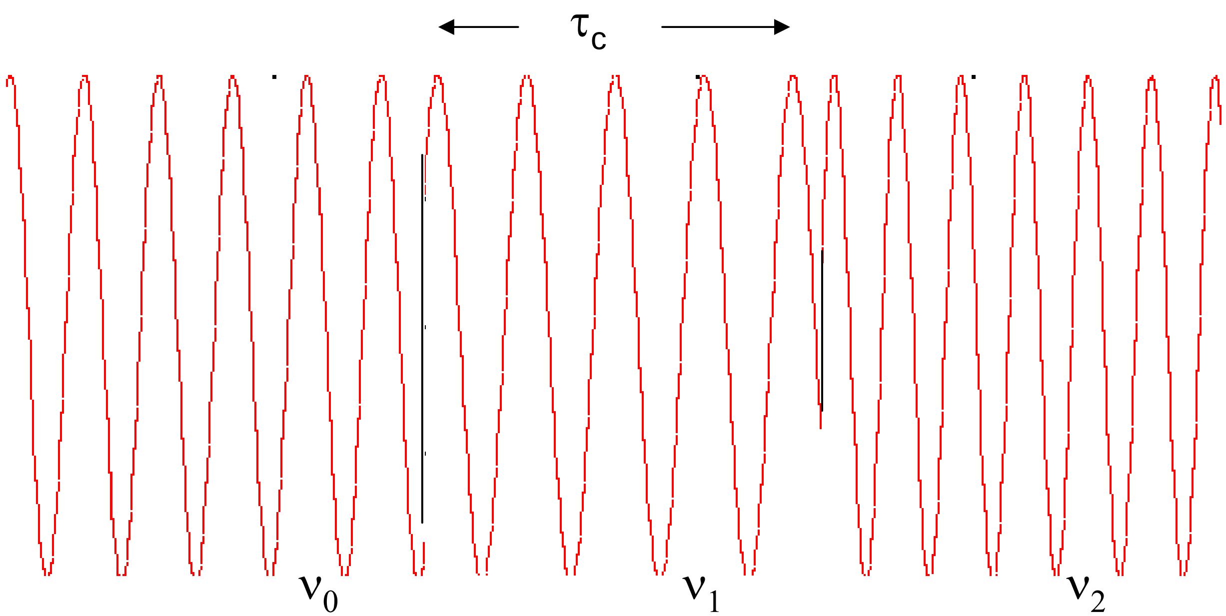
  (Gabor, Hanbury Brown, Born and Wolf, etc.)
- $E=V^r(t)$
- Assume polarized beam
- Assume beam is stationary – ensemble average = time average (e.g. no pulses!)

Coherence



Coherent

Ideal: fixed frequency
and phase



Partially Coherent

Band-limited frequency

Temporal Coherence

$$\tau_c \Delta \nu = 1$$

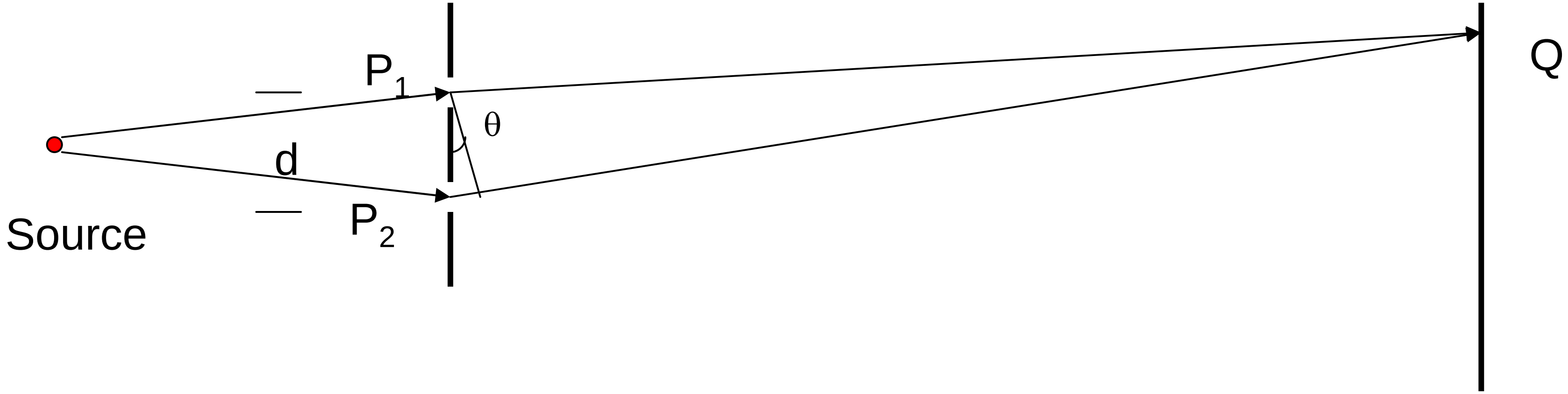
Over τ_c,wavefronts are equally spaced

Spatial Coherence – transverse coherence length, distance where wavefront is flat -> coherence area;

Longitudinal coherence = $c\tau_c$

2 Slit Experiment, 1 Detector

Young Double Slit Experiment

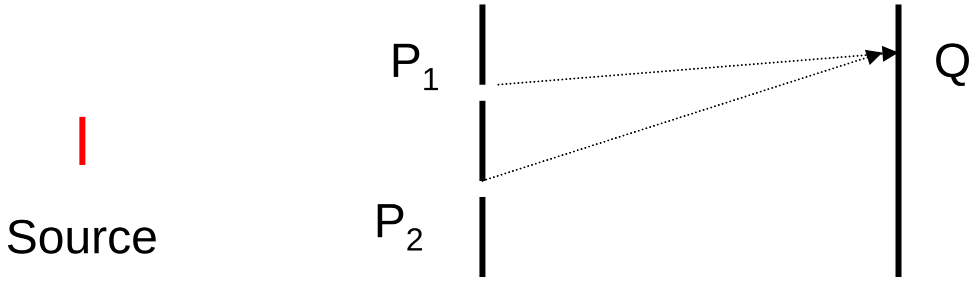


Interference condition for maxima: d $\sin(\theta) = m\lambda$

2 Slit Experiment, 1 Detector

(Hanbury Brown, Mandel and Wolf)

Two points P_1 and $P_{2,}$ are illuminated by a distant source. A detector is at a point Q on a screen



The field at Q is $V_Q(t) = V_1(t) + V_2(t+\tau)$, where τ is the time (path) difference for the radiation from the pinholes to Q

The (time average) intensity at Q is

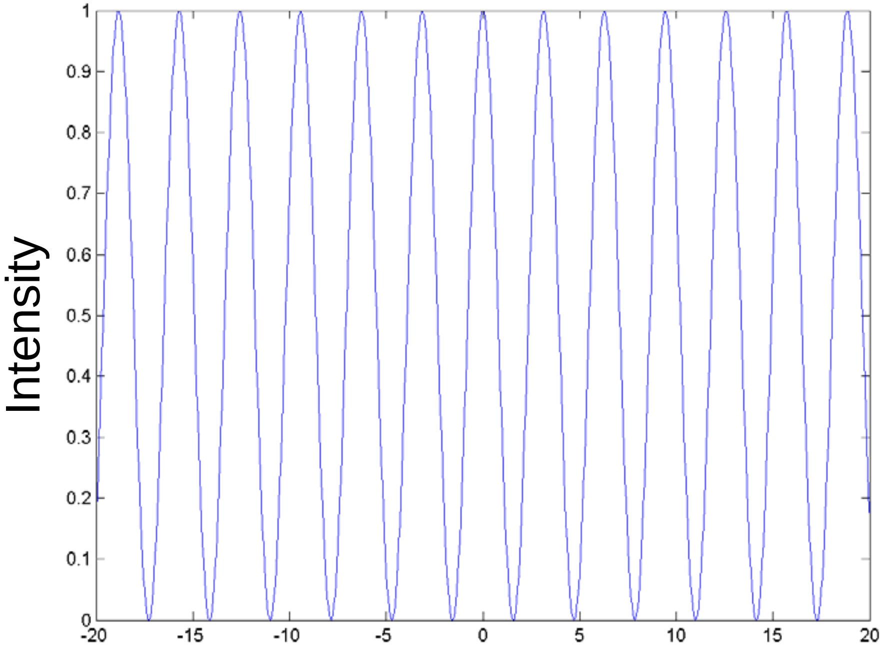
$I_Q = (1/2)\ I_1 + (1/2)I_2 + 2\ \mathrm{Re}[<V_1{}^*(t)V_2(t+\tau)>]$ (Young double slit expt)

cross correlation ~ interference term

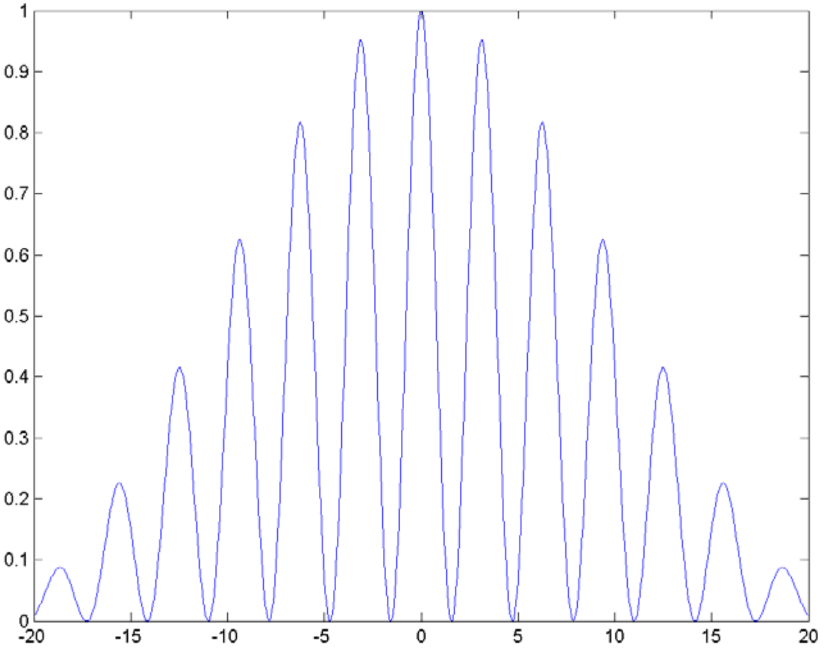
Extended Source

- A point source gives a simple interference pattern
- If the source has a finite size, the interference pattern is proportional to the Fourier transform of the intensity distribution over the source: the Van Cittert-Zernike theorem (Born and Wolf)
- This is similar to the Koonin-Pratt equation for heavy-ions (use first Born approximation for the 2 particle wavefunction)

Interference Patterns



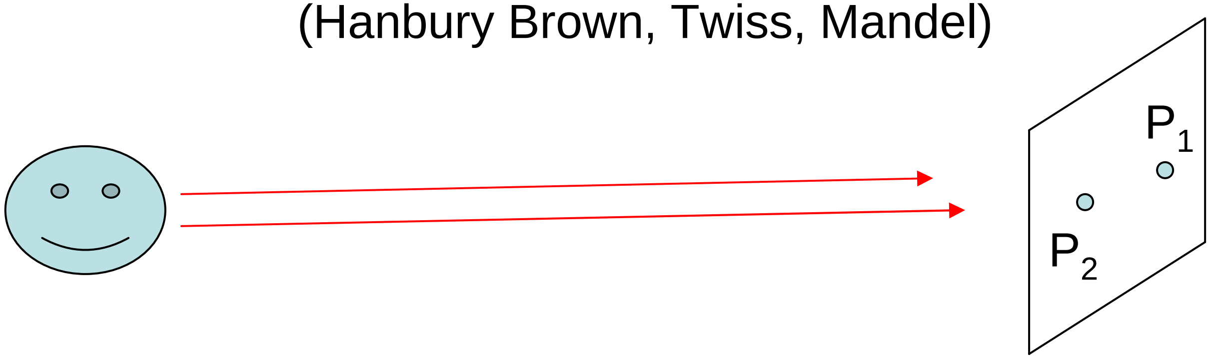
Point Coherent Source



Extended Source

Hanbury Brown Twiss Intensity Interferometry – Classical Description (simplified)

(Hanbury Brown, Twiss, Mandel)



An Intensity Interferometer measures the correlation between fluctuations of intensity at two separated points in partially coherent electromagenetic fields.

Two points P_1 and $P_{2,}$ illuminated by a distant source made up of independent radiators have intensities given by

$$I_1(t) = V_1{}^*(t)V_1(t)$$

$$I_2(t) = V_2{}^*(t)V_2(t)$$

The average of the intensities is

$$\langle I_1(t)I_2(t+\tau)\rangle = \langle V_1{}^*(t)V_1(t)V_2{}^*(t+\tau)V_2(t+\tau)\rangle$$

HBT theory, continued

$$\left\langle I_1(t)\, I_2(t+\tau)\right\rangle = \bar{I}_1 \bar{I}_2 + < \Delta I_1(t) \Delta I_2(t+\tau) >$$

$$\left\langle \Delta I_1(t)\, \Delta I_2(t+\tau)\right\rangle = \bar{I}_1 \bar{I}_2 \left| Fourier\ Transform\ of\ the\ source \right|^2$$

Note that there is no phase information.

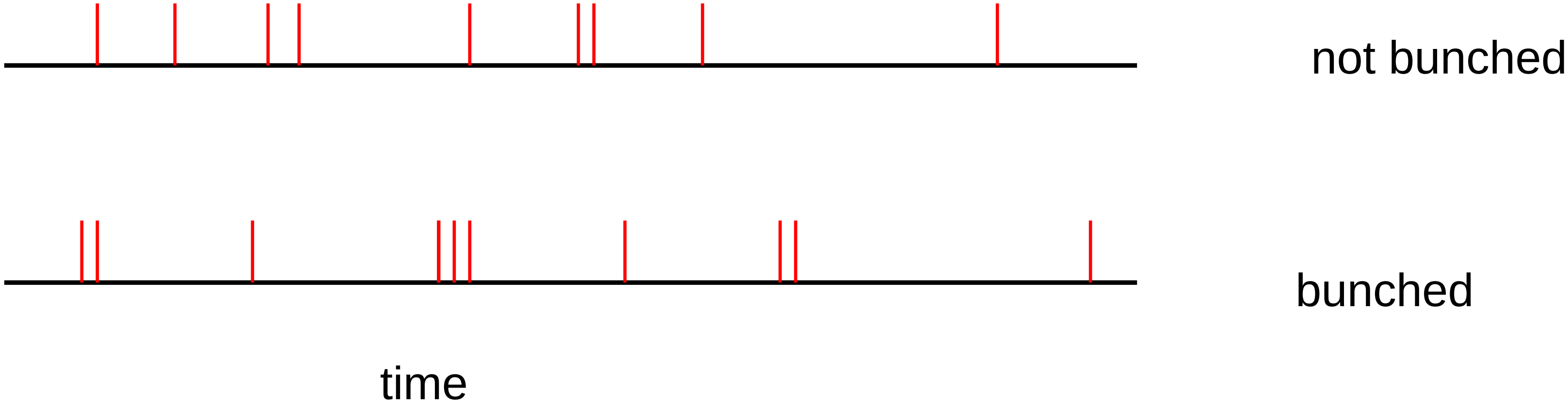
Quantum mechanically: measure counting coincidences between arrival times of individual photons. Bose-Einstein statistics implies photon bunching gives increased correlation; (Purcell)

Classical and QM results equivalent; statistics different; at high intensities non-linear optics likely to yield different physics (Glauber…)

See Crawford, Waves, for a simple example

Photon bunching

- Photons arrive in pairs
- Photon bunching in a single counter leads to photon bunching in a coincidence experiment
- Will lead to increase in correlation coefficient
- For HBT,quantum results basically same as classical, but statistical errors differ
- Random, electronic induced coincidences (shot noise)



Summary

- Thermal~Gaussian~Chaotic Source
- High Intensity (Irradiance)
- Narrow Bandpass (but Still Bright)
- Measure on a timescale ~$1/\Delta\nu$
- Light is now partially coherent
- Make lots of measurements, take average

Scattering

- Goldberger, Lewis, and Watson – “Use of Intensity Correlations to Determine the Phase of a Scattering Amplitude” (Phys. Rev, Dec 1963)
- They proposed extending Hanbury-Brown Twiss to the study of irradiated targets
- They use quantum quantum theory claim as opposed to the classical theory, quantum theory is “simpler and completely unambiguous” (implying classical theory is ambiguous)
- They suggest an important application may be to solve the phase problem in x-ray scattering
- ~origin of theory for 3-beam and reference beam x-ray studies

Goldberger,Lewis, and Watson proposal

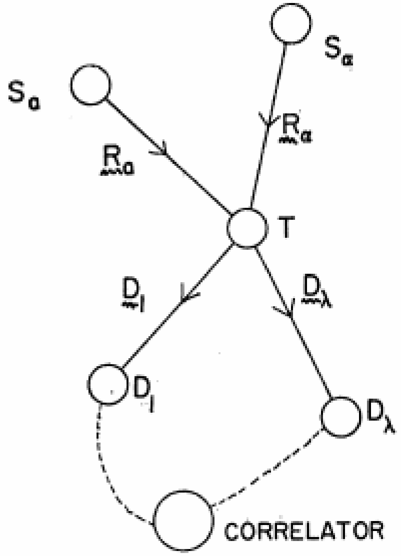
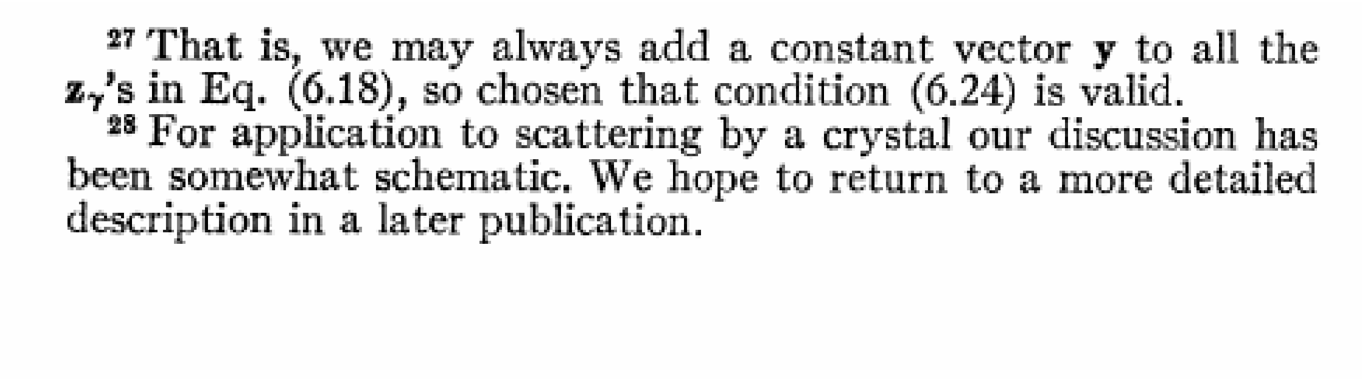


FIG. 3. Arrangement for measuring the correlated response of two detectors, from a target illuminated by two sources.

The idea of 2 sources seems to have mostly disappeared from the literature (?)

Followup paper never written?



[27] That is, we may always add a constant vector $\mathbf{y}$ to all the $\mathbf{z}_\gamma$'s in Eq. (6.18), so chosen that condition (6.24) is valid.

[28] For application to scattering by a crystal our discussion has been somewhat schematic. We hope to return to a more detailed description in a later publication.

Perhaps: “It’s solved, just fill in the details and find the money”

Gaussian Moment Theorem

- Higher order correlations give phase information
- In general, difficult to calculate
- It’s “simpler” for Gaussian variates; then all higher order correlations are expressible in terms of second-order correlations between pairs of variates (Mandel and Wolf, Mehta, Reed)
- If the distribution is not Gaussian, presumably you can solve the problem numerically

Highlights in Gaussian Moment Theory

- S.O. Rice "Mathematical Analysis of Random Noise, BSTJ v 24 1944 first(?) to come up with triple correlation formula
- Gamo H., Triple correlator of photoelectric fluctuations as a spectroscopic tool, Journal of Applied Physics 34, 875 (1963) (cites Rice and acknowledges a conversation with H.A. Gebbie at the Second Quantum Electronics Conference, Berkeley, 1961; see Gebbie in Advances in Quantum Electronics, J.R. Singer ed. 1961)
- T.S. Sato, S.Wadaka, J. Yamamoto, J.Ishii, Imaging system using an intensity triple correlator, Applied Optics 17 2047(1978) (cites Gamo) – imaging **incoherent** objects, 1d and 2d imaging of 100μm asymmetric objects with a laser and ground glass, and a lens to F.T. diffraction from the object
- A.S. Marathay, Phase function of spatial coherence from multiple intensity correlations, Proceedings of SPIE 628 273 (1966). Gives 3 and 4 photon correlation formulae (cites Gamo and others)
- P.R. Fontana, Multidetector intensity interferometers, JAPL, 54,473,1983

3 photon correlations for bosons

$$C_2 = 1 + \left|F_{12}\right|^2$$

$$\mathrm{C}_3 = 1 + \left|\mathrm{F}_{12}\right|^2 + \left|\mathrm{F}_{23}\right|^2 + \left|\mathrm{F}_{31}\right|^2 + \\ 2\,|\,\mathrm{F}_{12}\,||\,\mathrm{F}_{23}\,||\,\mathrm{F}_{31}\,|\,\underline{\cos(\phi_{12} + \phi_{23} + \phi_{31})}$$

This appears to be be computationally intensive; it's better to also do 4 photons (equations not shown)

Alternative phasing techniques

- Mandel – analyticity
- Gamo – three, four beams
- Triple correlations and bispectra
- Mendlovic et. Al. - Fractional triple correlations JOSA 15, 1658
- Holmes, Belen’skii – Cauchy Riemann equations JOSA A 21, 697
- None of the proposed phasing techniques appear to have been done with EM radiation (but see Sato)
- Are there others techniques, can the techniques be used in combination with each other?

Proposed experiments with synchrotron beams

- Ikonen - Interference Effects Between Independent Gamma Rays, PRL **68** 2759 (1992)
  - Proposed that photon bunching can be observed with highly monochromatic radiation
  - Two detectors
  - A synchrotron has high peak intensity with short pulses separated by detector dead time
  - Suggested that it would take about 10h of data collection to get a SNR of about 10 at ESRF, APS or SPRING-8 (then under construction)
  - Potential uses include energy width measurements and source-size determination
- Gluskin, McNulty, Vicarro and Howells – X-ray intensity interferometer for undulator radiation; NIM in Phys Res A319 p213 (1992)
  - Proposed to measure transverse coherences of an X-ray beam using HBT, use a linear multiplier (classical mode) ; 1300s

Accelerator X-ray beam physics

- Degeneracy factor δ – number of photons per phase space volume per coherence interval; alt. Number of photons per spatially and temporally coherent mode
- >>1 for optical lasers
- Often less than 1 for bright X-ray sources
- For undulators δ is $B\lambda^3/4c$ (~.0003 to .8)
- $\tau_c = \lambda^2/c\Delta\lambda = \lambda/c(\Delta\lambda/\lambda)$
- $l_c = \lambda/\Delta\theta = 2\pi/\Delta k$, measures source collimation / wave vector spread
- Electronic bandwidths typically much less than $1/\tau_c$, so temporal resolution of detector and electronics must be short

Experiments with Synchrotron Beams

- Gluskin, Alp, McNulty, Sturhan and Sutter. A classical Hanbury Brown-Twiss experiment with hard X-rays. *J. Synchrotron Rad.* (1999). **6**, 1065-1066

  2 Avalanche photodiode detectors (1.5ns time resolution, 15ns dead time)

  14.4 Mev photons, 5.5meV bandpass

  The measured spatial coherence area of the X-ray beam is in good agreement (15μm x 49 μm) with the prediction based on beam size.

  Data collection time was 12.8 hours

  Need to compare to their proposal

Experiments with Synchrotron Beams

- Yabashi,Tamasaku, and Ishikawa, Characterization of the Transverse Coherence of Hard Synchrotron Radiation by Intensity Interferometry. Phys. Rev. Lett. 87, 140801 (2001)
- Yabashi,Tamasaku, and Ishikawa, Measurement of X-Ray Pulse Widths by Intensity Interferometry. Phys. Rev. Lett. 88, 244801 (2002)
  - Spring 8
  - 14.4 keV photons, up to 120μeV bandpass, 25 and 27m undulators
  - 2 Avalanche photodiode detectors
  - Vertical beam size of 12.8 μm (.19μrad divergence)
  - Determined a beam pulse width of 32 ps

Experiments with Synchrotron Beams

- Kunimune,Yoda, Izumi, Yabashi, Zhang, Harami, Ando, and Kikuta. Two-Photon Correlations in X-rays from a Synchrotron Radiation Source. *J. Synchrotron Rad.* (1997). 4, 199-203

  14.4 keV photons at Tristan; Claimed to observe 2 photon correlations
- Tai, Takayama, Takaya, Miyahara, Yamamoto, Sugiyama, Urakawa, Hayano, and Ando. Chaotic nature of the stored current in an electron storage ring detected by a two-photon correlator for soft-x-ray synchrotron radiation. Phys. Rev. A 60, 3262–3266 (1999) ;Tai, Takayama, Takaya, Miyahara, Yamamoto, Sugiyama, Urakawa, Hayano, and Ando. A novel intensity interferometer for synchrotron radiation in the vacuum ultraviolet and soft x-ray regions. RSI 71 1256 (2000)

  Photon factory, KEK

  “preliminary”

  used a multiplier

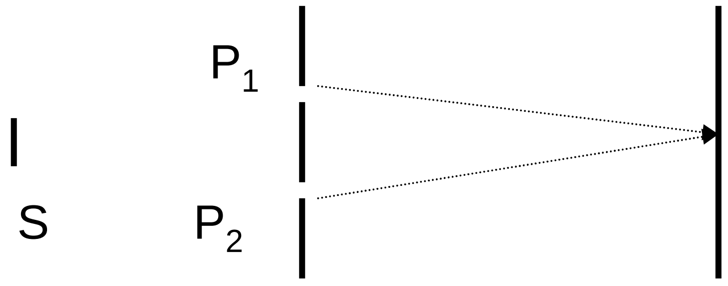
Some high energy/nuclear physics experiments

- Goldhaber, Goldhaber, Lee, and Pais. Influence of Bose-Einstein Statistics on the Antiproton-Proton Annihilation Process. Phys. Rev. 120, 300–312 (1960)The famous GGLP effect; Bevatron; pion production
- Zajc et al; Two-pion correlations in heavy ion collisions. Physical Review, C29:2173, 1984; (Bevalac)
- Frankel et al. Measurements of n-p correlations in the reaction of relativistic neon with uranium. Zeitschrift fur Physik A, 323:391 1986; (Bevalac)
- Aggarwal et al. Three-Pion Interferometry Results from Central *Pb*+*Pb* Collisions at 158*A GeV/c* Phys. Rev. Lett. 85, 2895–2899 (2000) (CERN)
- Adams et al. Three-Pion Hanbury Brown–Twiss Correlations in Relativistic Heavy-Ion Collisions from the STAR Experiment. Phys. Rev. Lett. **91**, 262301 (2003)

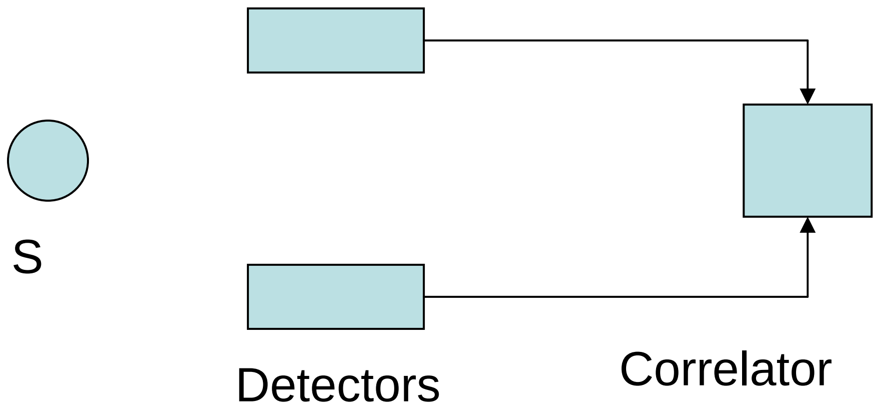
Techniques for coincidence counting and analysis for 3 bosons or fermions has been worked out

Only problem is: you can’t really verify your result (results difficult to interpret or non-interpretible), in molecular structure we can!

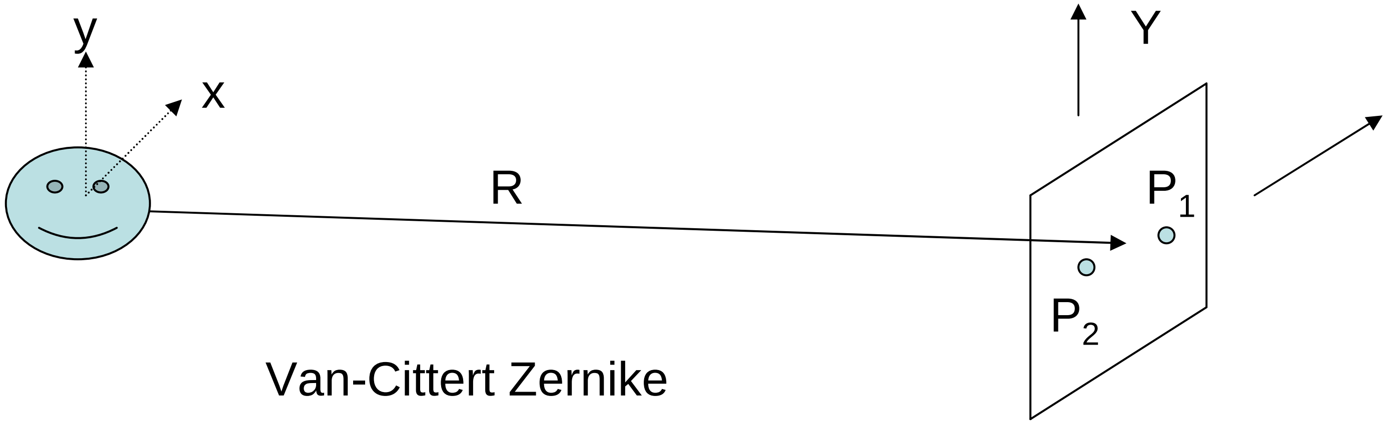
“Review of Experiments”



Young double slit expt.

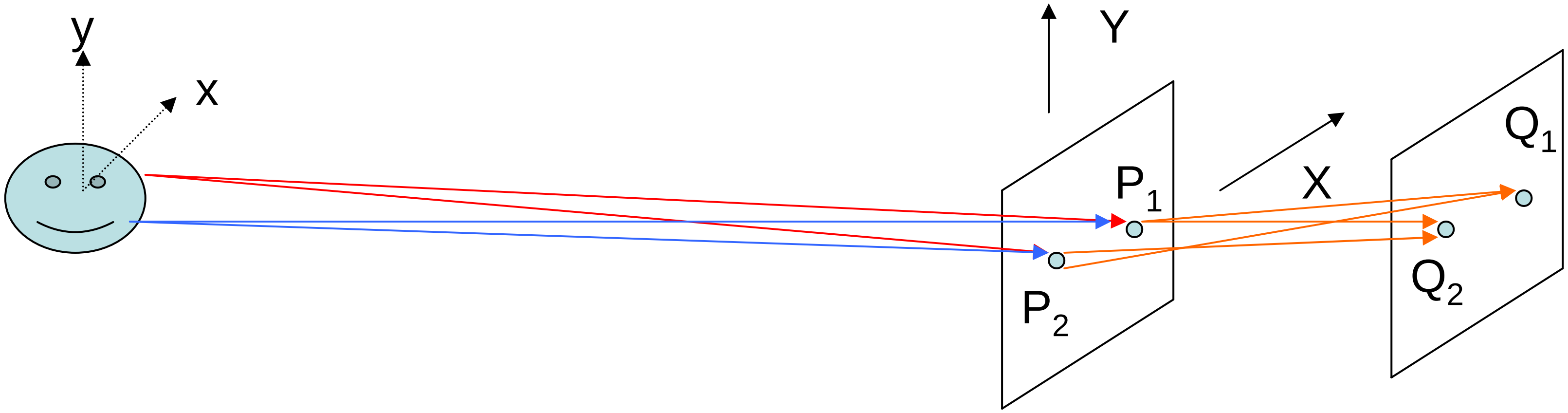


HBT



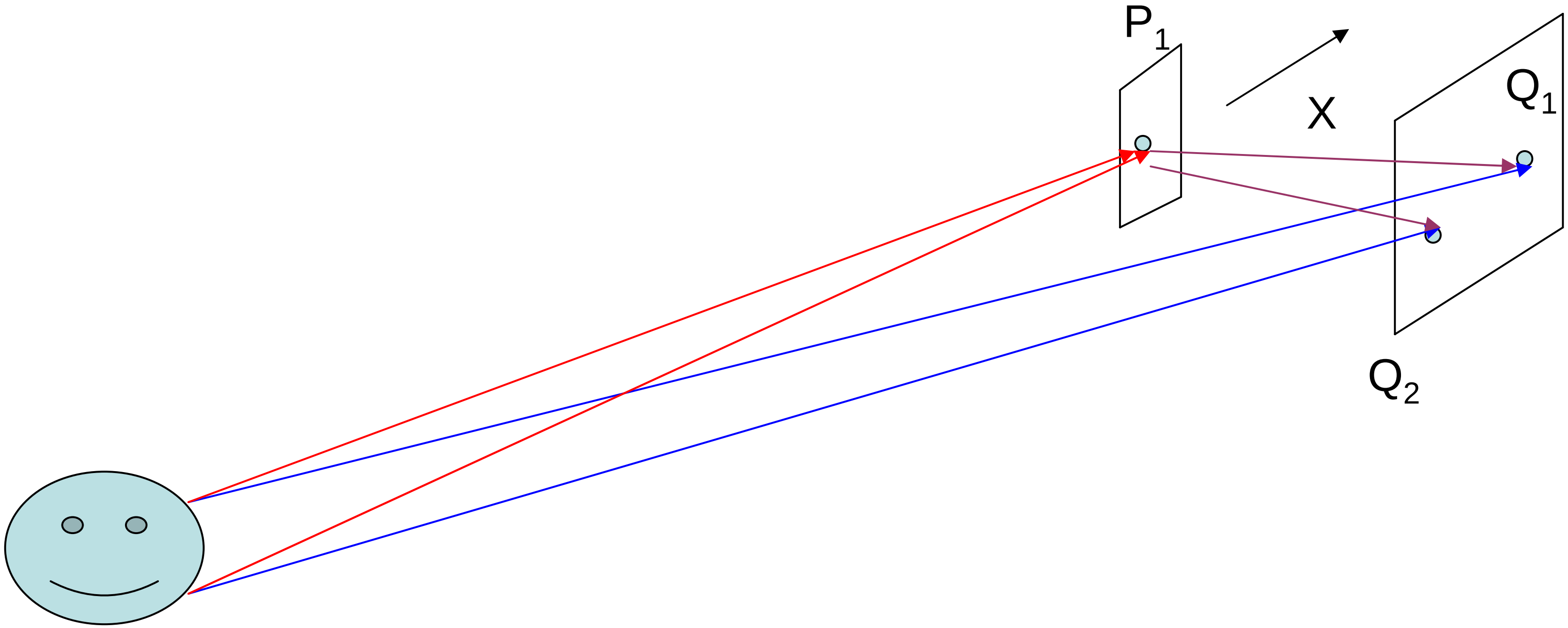
Van-Cittert Zernike

Simplest imaging experiment



Alternative simplest imaging experiment

Get phase information?



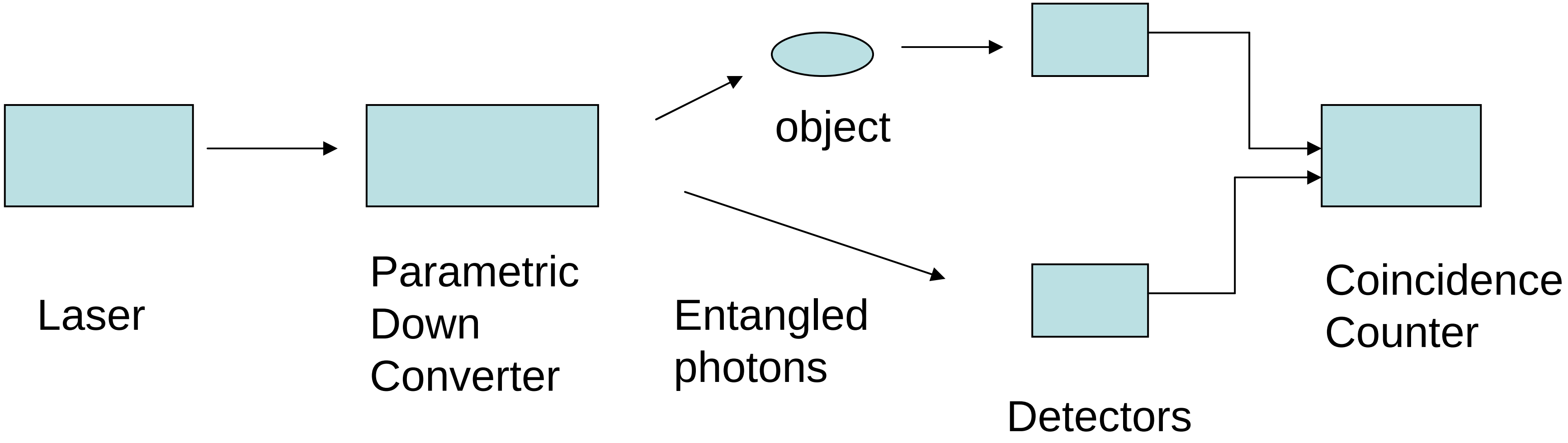
This appears to still be controversial!

Imaging Experiments:
Why weren’t they done sooner?

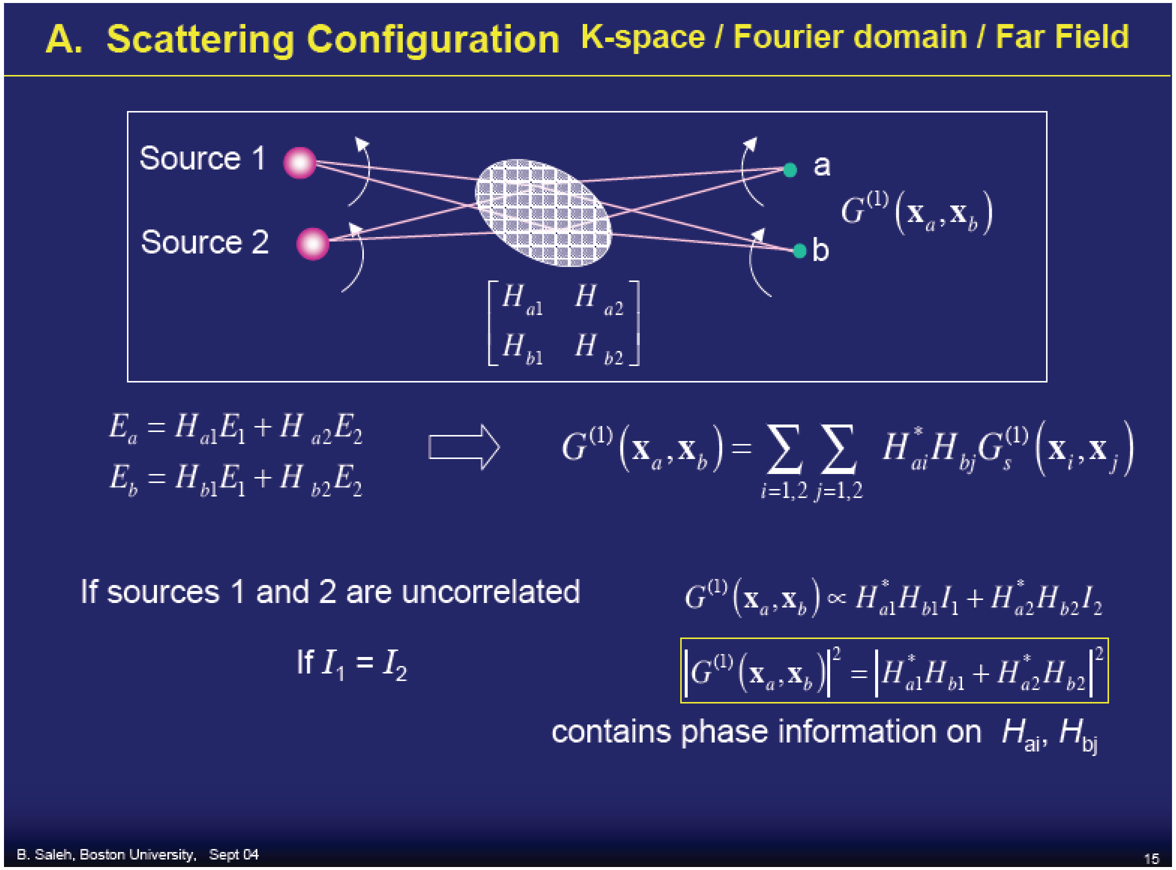
- Maybe it was and we can’t find it
- Too easy?
- Too hard?
- No one thought of it?
- It’s obvious?
- Maybe it was done, and we just don’t understand the paper (Sato 1977, but they also had to use a lens)
- Simulated vs real experiments?
- Classical and quantum imaging theory seem’s to be worked out (see Goodman on Schell’s theorem)
- By the time imaging was being done there seems to be an entangled mess of entanglement; then had to untangle them!
- See Trebino, Gustafson, Siegman “Fourth-order partial-coherence effects in the formation of integrated-intensity gratings with pulsed light sources” J.Opt. Soc. Am. B. **3** 125 (1986) and Ebstein, JOSA 8 p 1442 1991, 4th order correlation interferometry; transmission object is part of the speckle generator

Ghost Imaging Experiments

- Use entangled photons, (later found that thermal photons work but with worse S/N);
- 1 photon through the object, 1 photon is a reference beam, need a lens
- Not sure if this is of any interest to us



Saleh suggests 2 photon imaging may have phase information. (http://web.bu.edu/qil/pdf/Saleh-Tutorial-Corsica-0904.pdf)



A. Scattering Configuration K-space / Fourier domain / Far Field

$G^{(1)}(\mathbf{x}_a,\mathbf{x}_b)$

$$\begin{bmatrix} H_{a1} & H_{a2} \\ H_{b1} & H_{b2} \end{bmatrix}$$

$$E_a = H_{a1}E_1 + H_{a2}E_2$$
$$E_b = H_{b1}E_1 + H_{b2}E_2$$

$$\Rightarrow \quad G^{(1)}(\mathbf{x}_a,\mathbf{x}_b) = \sum_{i=1,2}\sum_{j=1,2} H^*_{ai}H_{bj}G^{(1)}_s(\mathbf{x}_i,\mathbf{x}_j)$$

If sources 1 and 2 are uncorrelated $G^{(1)}(\mathbf{x}_a,\mathbf{x}_b) \propto H^*_{a1}H_{b1}I_1 + H^*_{a2}H_{b2}I_2$

If $I_1 = I_2$ $\boxed{\left|G^{(1)}(\mathbf{x}_a,\mathbf{x}_b)\right|^2 = \left|H^*_{a1}H_{b1} + H^*_{a2}H_{b2}\right|^2}$

contains phase information on H_{ai}, H_{bj}

Classical Coherence Imaging and Quantum Two-Photon Imaging B. E. A. Saleh
First International Workshop: *Imaging at the Limits*, IESC, Cargèse, Corsica (September 2004)

This looks like the Goldberger,Lewis and Watson proposal (He also cites GLW).

Can we design a viable experiment?

- Typically, the design of correlation experiments requires an extensive interaction between theorists and experimenters.
- We need the help of theorists who understand both optics and the needs of structural biologists!
- The field has been developed by scientists working in classical optics, imaging, quantum optics, astronomy, high energy physics, and nuclear physics – We need to get people to speak the same language!
- Literature is wrong or vocabulary changes or may appear to change (e.g. light from a Hg lamp described as coherent is probably partially coherent)
- “Assume” Mandel and Wolf (and Born and Wolf) did the “correct” classical theory, Glauber did the “correct” quantum theory, Loudon for an “understandable” quantum theory, Goodman for statistical optics – rest is details
- We need a preliminary design; one mistake and all can FAIL!
- Aside from random coincidences, data should have low background
- Need only first approximation to phases, refine after that
- Both the accelerator and detector system are likely to be extremely expensive

First iteration design for a multi-photon measurements for protein crystallography

- Thermal (or pseudo-thermal) X-ray light source (problems with beam being stationary?)
- Narrow pulses (~ 1 fs or better (???) unless someone invents really fast electronics) (currently technologically difficult); Can a pulsed laser help shorten the electron beam pulse? Rotating ground glass only good to ~10^{-5} seconds?
- The pulses need to be separated by the detector read out and recovery time
- Small x-ray beam divergence
- Polarized X-rays simplifies the analysis

Experimental design (continued)

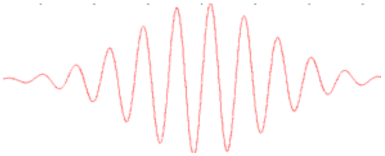
- Single molecule or small (<25 μ) crystals
- Photon energy should be about 6 keV
- About 10^7 monochromatic photons in each pulse ($\Delta E/E <\sim 1$meV) (necessary if pulse is short?)
- Detector system consists of hundreds of individual detectors (~APDS; expensive)
- Hardware multiplier/correlator (FPGA, ASIC)?
- Other types of correlation measurements may yield useful information (studying Raman, resonance fluorescence, etc); Ofir claims FCS is analogous to triple correlation measurements
- Will nonlinear phenomena help?

Is Intensity Interferometry/Photon Correlation Measurement Viable?

- Is the physics right – both theoretically (probably) and experimentally? (given infinite resources -possibly – but there are potential problems in the timing requirements)
- Should we run in current multiplier mode, or correlation mode?
- Short pulses “induce” coherence; need partial coherence
- Can a real source and real detector be built that can do the experiment?
- Can a source and detector be built for a finite price?
- Can a source and detector be built for a reasonable price and can the experiment be done in a reasonable time? (compare to properly done MAD experiments)
- Will the number of photons required be less than required for MAD? (and therefore less radiation damage)
- Can be used when protein resistant to MAD

Some potential problems

- For a pulsed source, the beam isn’t stationary and ensemble averages aren’t equal to time averages
- For real detectors, measurement time T is greater than coherence time τ_c, the correlation is reduced by τ_c/T
- Propagation through volume source is more complicated than a plane source (Beran and Parrent) (but compare to Goldberger?)
- Treat beam as a combination of wavepackets?



- What is the proper source size and can we make it?
- Issues with T~ τ_c
- Simulations may help

Experiments to consider

- Pulsed laser experiments and classical HBT measurement
- Use a pulsed laser experiment to do Young slits and diffraction grating experiments, reconstruct the scatterer
- Try an experiment at an accelerator (APS, SPRING-8, ESRF, ALS – soft X-ray)
- Experiments at a next generation X-Ray light source – initially just need 3 detectors, then build a system with more detectors ($$$)

To do

- Refine design parameters
- Simulate if statistical optics doesn’t work (and even if it does!)
  we have fast computers now;
  warning: correlations more complicated than single particle simulations)
- Get help/criticism (Very few “qualified” people”)
- Reconcile quantum and classical theory
  Schell’s theorem, quantum counterpart; field theory view; timing issues
- Do some experiments

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