Entangled State Experiments using Photon Pairs

SQuInT retreat lectures, June '01.

Daniel James (LANL)

– all rights reserved –

- Entangled States: pair of two-level systems.
  - particle spins
  - flux quanta
  - atomic / ion quantum states / quantum dot
  - photon degrees of freedom:
    - ○ modes
    - ○ photon number
    - ○ frequency
    - ○ POLARIZATION * — the one we are most interested in!

Quantum Theory started with optics 100 years ago and its still leading the charge!

$$|\Psi_1\rangle = a|H\rangle + b|V\rangle$$

two "qubits":

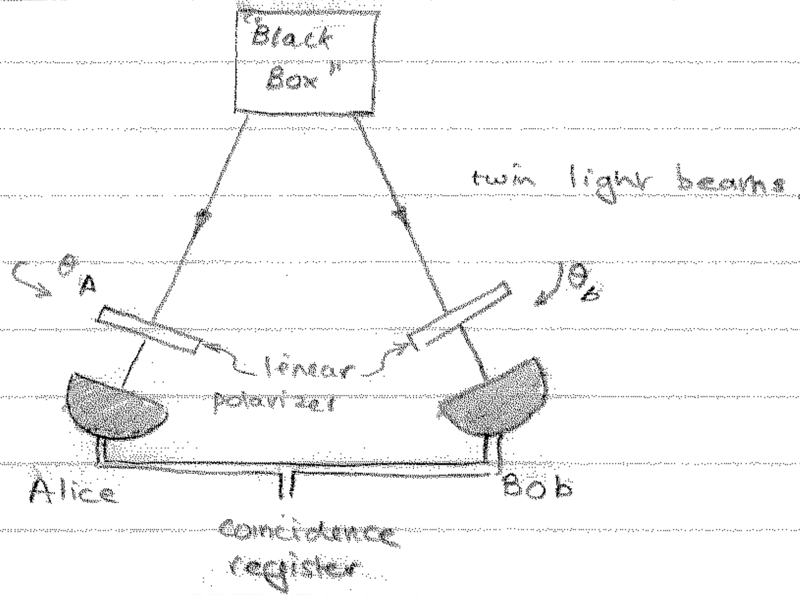
$$|\Psi_2\rangle = a|HH\rangle + b|HV\rangle + c|VH\rangle + d|VV\rangle$$

$$\neq \underbrace{(A|H\rangle + B|V\rangle) \otimes (C|H\rangle + D|V\rangle)}_{\text{separable state}}$$

– "I would not call [entanglement] *one*, but rather *the* characteristic trait of quantum mechanics..."

– Erwin Schrödinger

Example : classical versus Quantum Correlation

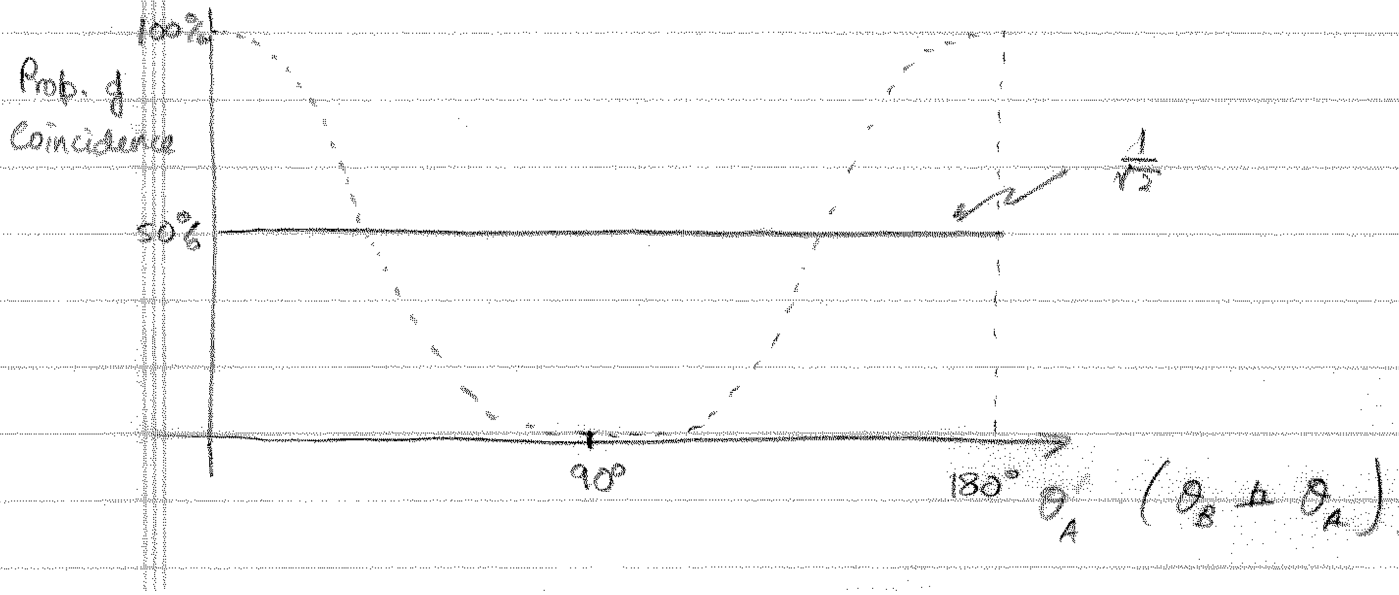


Case 1 : classical correlations :

every photon that comes to Alice is $|H\rangle$
every photon " " " Bob " $|V\rangle$

combined state : $|HV\rangle$

Alice's Polarizer	Bob's Polarizer	% Coincidences.
$|H\rangle$	$|V\rangle$	100%
$|D\rangle$ (45°)	$|\bar{D}\rangle$ (135°)	50%
$|H\rangle$	$|V\rangle$	0%



Case 2 : quantum correlations

superposition state

$$|\Psi_-\rangle = \frac{1}{\sqrt{2}}\left(|HV\rangle - |VH\rangle\right)$$

i.e. A gets a $|H\rangle$, B a $|V\rangle$ or A gets a $|V\rangle$ B a $|H\rangle$ with equal probability amplitudes

In this case coincidence percentage is always 50%.

> Homework problem: try to find a quantum state which is not entangled but which reproduces this result !

Creation of Entangled photon pairs

what are "photons"

Free, classical field

$$\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\underline{E}(r,t) = 0$$

$$\nabla . \underline{E}(r,t) = 0$$

decompose into a set of spatial modes:

$$\underline{E}(r,t) = \sum_\lambda \underline{\Phi}_\lambda(\underline{r})\,\alpha_\lambda(t) + c.c.$$

$$\left.\begin{aligned}(\nabla^2 + \omega_\lambda^2)\,\underline{\Phi}_\lambda(r) &= 0\\ \nabla . \underline{\Phi}_\lambda &= 0\end{aligned}\right\}$$ examples: plane waves $\underline{\Phi}_\lambda = e_\lambda \frac{1}{\sqrt{V}} e^{i\underline{k}_\lambda \cdot}$ (Planck, Dirac)

laser modes, etc.

$$\int \underline{\Phi}_\lambda(r) . \underline{\Phi}_\mu(r)\, d^3r = \delta_{\lambda\mu}$$

— Mode amplitude —

$$\boxed{\frac{d^2}{dt^2}\alpha_\lambda(t) + \omega_\lambda^2\,\alpha_\lambda(t) = 0.}$$

Simple Harmonic oscillator

Quantum theory of fields: replace this classical SHO with a quantum one! (P.A.M. Dirac)

$$c_\lambda(t) \rightarrow \hat{c}_\lambda(t) = i\sqrt{\frac{\hbar}{2\omega_\lambda \epsilon_0}}\, \hat{a}_\lambda^\dagger(t)$$

field operator

$$\hat{\underline{E}}(r,t) = \sum_\lambda i\sqrt{\frac{\hbar}{2\omega_\lambda \epsilon_0}}\, \underline{\Phi}_\lambda(r)\, \hat{a}_\lambda^\dagger(t) \quad + \text{ h.a.}$$

Observable quantities (fields, field correlation functions etc) can be calculated if you know the state of the field

E.g. two modes, using number state representation

$$|\Psi\rangle = c_{00}|0,0\rangle + c_{0,1}|0,1\rangle + c_{0,2}|0,2\rangle + \ldots$$
$$+ c_{1,0}|1,0\rangle + c_{1,1}|1,1\rangle + \ldots$$
$$+ c_{2,0}|2,0\rangle + \ldots \qquad = \sum_{n,m} c_{nm}|n,m\rangle$$
$$\vdots \qquad \text{etc.}$$

$c_{n,m}$ = amplitude associated with n 'photons' in first mode + m 'photons' in the second mod

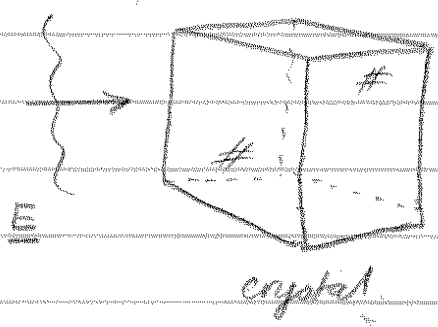
- infinite numbers of modes to play with.
- we use two spatial modes (each with one photon) plus the two polarization degrees of freedom for each mode.

field dynamics:

- Electromagnetic energy $U = \int d^3r \left(\frac{\epsilon_0}{2}E^2 + \frac{\mu_0^2}{2}B^2\right)$

$$\rightarrow \sum_\lambda \hbar\omega_\lambda\left(\hat{a}_\lambda^\dagger \hat{a}_\lambda + \frac{1}{2}\right)$$

Non-linear Interactions



Polarization of the crystal

$$P_i = \chi_{ij}^{(1)} E_j + \chi_{ijk}^{(2)} E_j E_k + \ldots$$

$$\text{Energy} = \int \underline{E} \cdot \underline{P} \, dV$$

$$= \sum_{\lambda\mu\nu} \eta_{\lambda\mu\nu} \hat{a}_\lambda^\dagger \hat{a}_\mu^\dagger \hat{a}_\nu + \text{h.a.} \quad (+ \text{ other terms} \ldots)$$

— 3 wave mixing.

ν photon in creates λ, μ photons out.

Phase matching considerations: $\eta_{\lambda\mu\nu}$ is small unless:

$\omega_\lambda + \omega_\mu = \omega_\nu$. (conservation of energy)

— requires interaction picture —

$\underline{k}_\lambda + \underline{k}_\mu = \underline{k}_\nu$ (conservation of momentum)

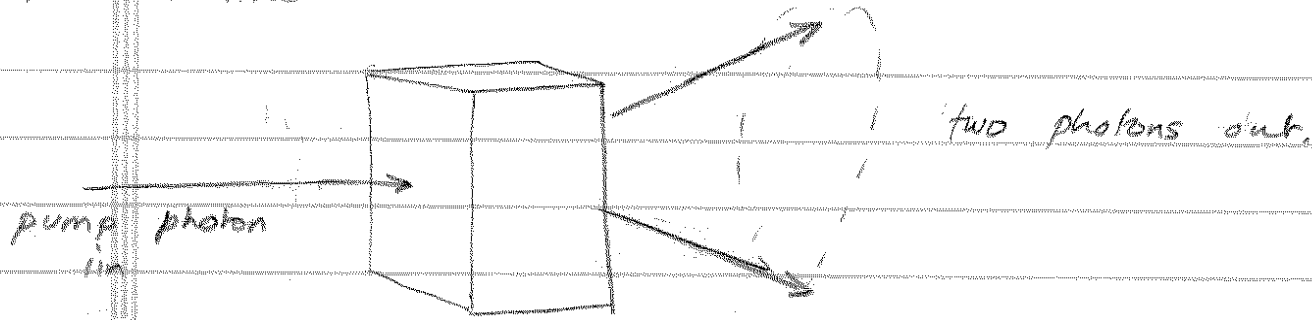
— plane wave modes.

- use bi-refringence to meet the momentum condition.
- analysis gets REAL complicated...

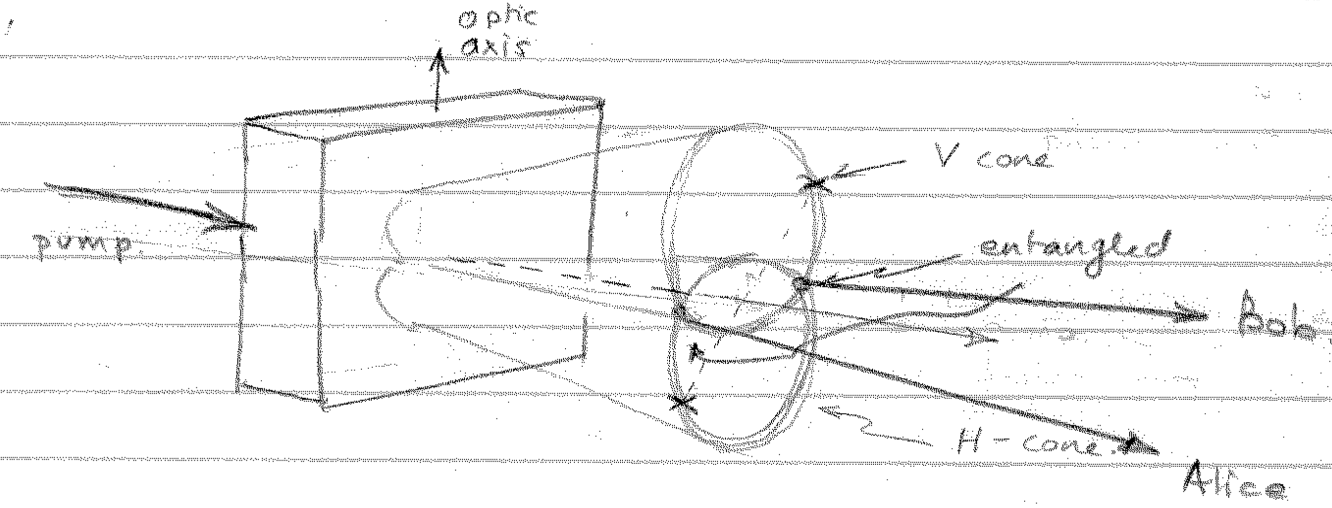
type I phase matching: μ, ν: $\parallel$ polarization

type II " " : μ, ν: $\perp$ polarization

Bottom line:

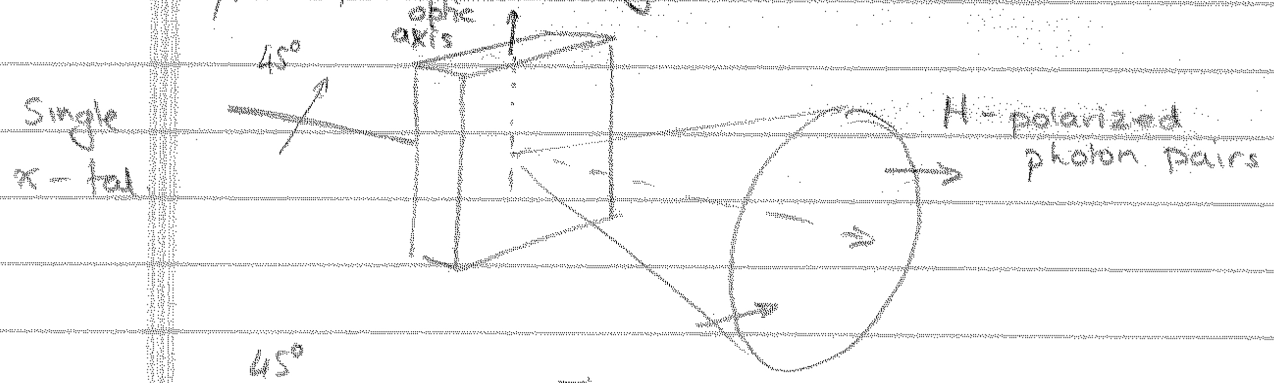


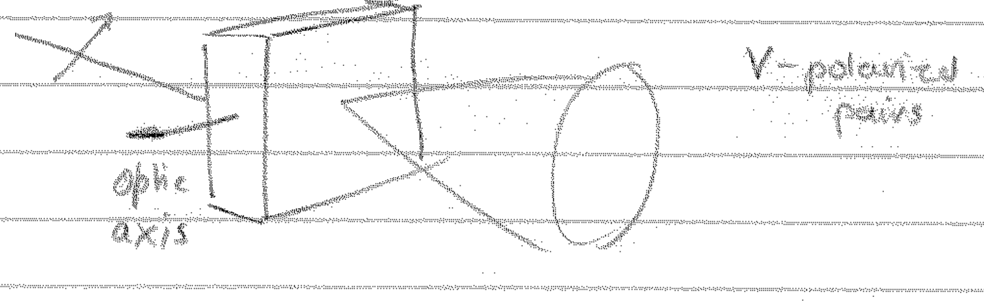
- circular symmetry : cones of possible photon emissions.



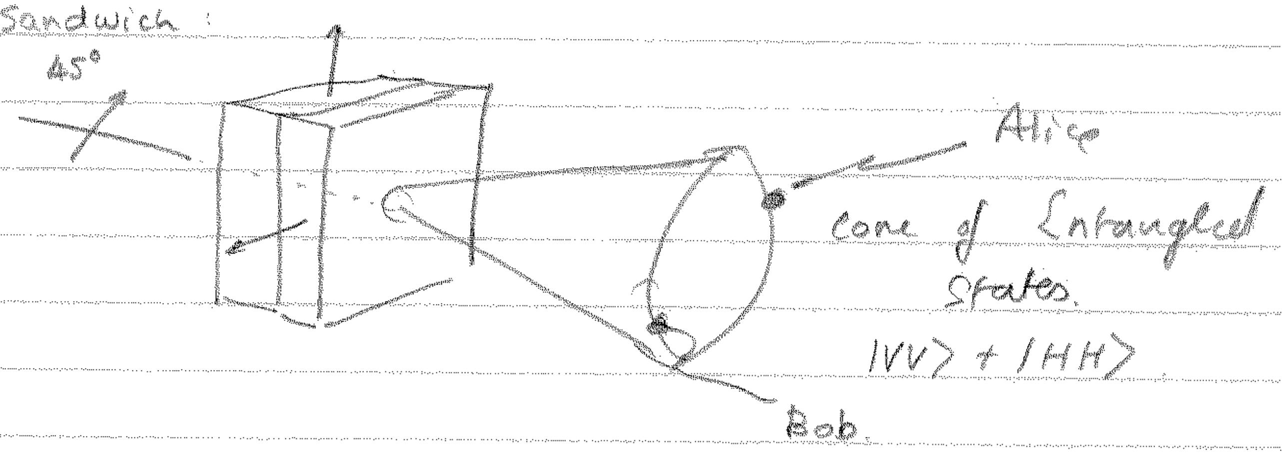
overlap region : $|H\rangle|V\rangle + |V\rangle|H\rangle$

type I phase matching.





Sandwich :



Note on post - section & the vacuum

The actual States that are being produced are

$$\exp\left[i\eta\left\{\hat{a}_H^{+}\,\hat{b}_H^{+}\,\hat{c}_V + \hat{a}_V^{+}\,\hat{b}_V^{+}\,\hat{c}_H + \text{h.a.}\right\}\right]|\text{vac}\rangle|\text{vac}\rangle|\text{pump state}\rangle$$

creation of H polarized photon for Alice

creating H polarization photon for Bob

anihilation of a V-polarization pump photon

$$= |\text{vac}\rangle|\text{vac}\rangle|\text{pump}\rangle$$

$$i\eta\left\{|H\rangle|H\rangle\,\hat{c}_V|\text{pump}\rangle + |V\rangle|V\rangle\,\hat{c}_H|\text{pump}\rangle\right\}.$$

$$+ O(\eta^2)$$

trace over pump:

coefficients (can be random if pump is in a mixed State)

$$|\text{vac}\rangle|\text{vac}\rangle + i\eta\left\{\alpha|HH\rangle + \beta|VV\rangle\right\} + O(\eta^2)$$

— coherent superpositions.

i.e. we have a whole heap of *nothing* (vacuum)
+ a small amplitude for an entangled state.

- vacuum states have no observable effect here.
- i.e. Random emission of photons
  "post selected states"
  - non-deterministic creation of entanglement.

with this caveat, we *assert* that the state of the "post-selected" photon pair is
$|\Psi_2\rangle = \alpha|HH\rangle + \beta|VV\rangle$.
where $|\alpha|^2+|\beta|^2=1$ and α, β can be controlled by changing the state of the pump.

Mixed States.

pure state of one qubit $|\Psi_1\rangle = \alpha|0\rangle+\beta|1\rangle$.

deterministic

mixed state: Random numbers.
characterize these random numbers by
i) their means $\overline{|\alpha|^2}$, $\overline{|\beta|^2}$
ii) their cross-correlation $\overline{\alpha\beta^*} = (\overline{\alpha^*\beta})^*$.

Represent state as a *density operator*

$$\hat{\rho} = \overline{|\Psi_1\rangle\langle\Psi_1|}$$

or, in matrix form,

$$\rho_{nm} = \begin{pmatrix} |\alpha|^2 & \alpha\beta^* \\ \alpha^*\beta & |\beta|^2 \end{pmatrix} \quad \text{— density matrix.}$$

- this is the most you can know about the state.

Rotate these waveplates : $\begin{pmatrix} |H\rangle \\ |V\rangle \end{pmatrix}$ basis

$$\hat{U}_{QWP}(\theta) = \frac{1}{\sqrt{2}} \begin{pmatrix} i - \cos(2\theta) & \sin(2\theta) \\ \sin(2\theta) & i + \cos(2\theta) \end{pmatrix}.$$

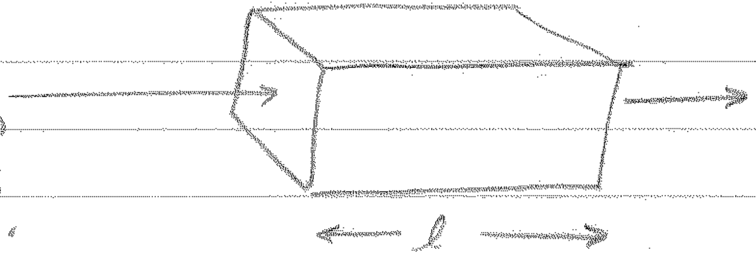
$$\hat{U}_{HWP}(\theta) = \begin{pmatrix} \cos(2\theta) & -\sin(2\theta) \\ -\sin(2\theta) & -\cos(2\theta) \end{pmatrix}.$$

— single qubit operations are relatively straightforward with photons.

------ Break here ---------

"Decoherence" : Random phase lag between $|H\rangle$ and $|V\rangle$ — can be done with a simple birefringent crystal.

goes after 11.



$k(n_e - n_o)L \gg$ Longitudinal coherence length of photons.

— this is not <u>really</u> decoherence because it can be reversed.

10½

Lecture 2

Reprise:

i) Creation of polarization entangled states of pairs of photons by down-conversion

$$\delta H(t) = \underline{P}.\underline{E} =$$

$$\chi_{ijk} E_i E_j E_k = \sum_{\lambda\mu\nu} \left(i \sqrt{\frac{\hbar}{2\epsilon_0}} \right)^3 \frac{1}{\sqrt{\omega_\lambda \omega_\mu \omega_\nu}} \quad \chi_{ijk} \int \Phi_{\lambda i}(r)\, \Phi_{\mu j}(r)\, \Phi^*_{\nu k}(r)\, dV$$

$$\times \; \hat{a}_\lambda^{\dagger} \hat{a}_\mu^{\dagger} \hat{a}_\nu \quad \underbrace{\exp i\,(\omega_\lambda + \omega_\mu - \omega_\nu)t}_{\text{(Interaction picture)}} \qquad + \text{h.a.}$$

(+ other terms)

$$|\Psi_{out}\rangle = \hat{U}\,|\Psi_{in}\rangle$$

$$\hat{U} = \mathrm{T} \exp\left\{ \frac{-i}{\hbar} \int_{-\infty}^{\infty} \delta H_I(t)\, dt \right\}.$$

$$\approx \quad \hat{\mathbb{I}} + \sum_{\lambda\mu\nu}{}' \eta_{\lambda\mu\nu}\, \hat{a}_\lambda^{\dagger} \hat{a}_\mu^{\dagger} \hat{a}_\nu \quad + \text{h.a.}$$

limited sum:

$$\omega_\lambda + \omega_\mu - \omega_\nu = 0$$

$$\underline{k}_\lambda + \underline{k}_\mu - \underline{k}_\nu = 0.$$

ii) Two types of down-conversion entangled sources: Type II ("figure 8") and Type I (Sandwich)

iii) Control created state by: (a) state of pump beam.

(b) local unitaries

(c) decoherence stage.

(see previous)

Measurement of States.

A simple example of "quantum state tomography"

tomos - Gk. for "slice".

- measurement of 3-D objects by measuring a bunch of "slices". (Computer Tomography Scans)
- Applied to measurement of Wigner function of Harmonic oscillator state (Raymer) - math same as CT scans hence "Quantum state Tomography"
- come to represent all quantum state estimation.

- deal with in some detail since it will apply to all quantum computers.

Simple case: 1 - photon beam. (Stokes, 1852)

- same Stokes as Stokes' theorem $\iint_S (\nabla \times \underline{A}).\underline{d\sigma} = \oint \underline{A}.\underline{dl}$.

(which was first "published" in a finals examination).

Measure complete polarization state of a photon by 4 measurements

a) with a filter that transmits ALL polarizations (I_0)

" $= \hat{\Pi}_0 = |H\rangle\langle H| + |V\rangle\langle V| = \hat{\mathbb{1}}$

b) with a Horizontally oriented linear polarizer $(I_1)\ \hat{\Pi}_1 = |H\rangle\langle H|$

c) " " diagonally " " " $(I_2)\ \hat{\Pi}_2 = |\bar{D}\rangle\langle\bar{D}|$

d) " " Right - circular " $(I_3)\ \hat{\Pi}_3 = |R\rangle\langle R|$

use $|R\rangle, |L\rangle$ basis:

$$\begin{pmatrix} |R\rangle \\ |L\rangle \end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & -i \\ 1 & +i \end{pmatrix} \begin{pmatrix} |H\rangle \\ |V\rangle \end{pmatrix}$$

Stokes parameters:

$$S_0 = I_0 = \mathcal{N}\left(\langle R|\hat{\rho}|R\rangle + \langle L|\hat{\rho}|L\rangle\right) = \mathcal{N}$$

$$S_1 = 2I_1 - I_0 = \mathcal{N}\left(\langle R|\hat{\rho}|L\rangle + \langle L|\hat{\rho}|R\rangle\right).$$

$$S_2 = 2I_2 - I_0 = i\mathcal{N}\left(\langle R|\hat{\rho}|L\rangle - \langle L|\hat{\rho}|R\rangle\right)$$

$$S_3 = 2I_3 - I_0 = \mathcal{N}\left(\langle R|\hat{\rho}|R\rangle - \langle L|\hat{\rho}|L\rangle\right).$$

constant
(beam intensity, detector efficiency, etc.)

$\hat{\rho}$ = density matrix for the single photon

$$\boxed{\hat{\rho} = \frac{1}{2}\sum_{\mu=0}^{3} \frac{S_\mu}{S_0}\,\hat{\sigma}_\mu}$$

Pauli matrices in the $|R\rangle, |L\rangle$ basis:

$$\hat{\sigma}_0 = |R\rangle\langle R| + |L\rangle\langle L|$$

$$\hat{\sigma}_1 = |R\rangle\langle L| + |L\rangle\langle R|$$

$$\hat{\sigma}_2 = i\left(|L\rangle\langle R| - |R\rangle\langle L|\right)$$

$$\hat{\sigma}_3 = |R\rangle\langle R| - |L\rangle\langle L|$$

- S_μ are not the outcomes of measurements : $S_\mu \not\propto \mathrm{Tr}\{\hat{\rho}\,\hat{\Pi}\}$

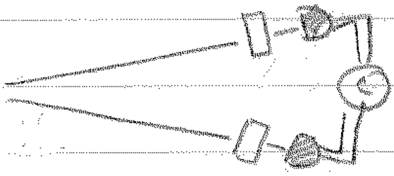
Now extend this idea to 2-qubits

$$\hat{\rho} = \frac{1}{4} \sum_{\mu,\nu=0}^{3} r_{\mu\nu} \, \hat{\sigma}_\mu \otimes \hat{\sigma}_\nu$$

$\hat{\sigma}_0 = \hat{\mathbb{I}}$

$\hat{\sigma}_i$ = pauli matrices.

normalization $r_{00} = 1$



Take <u>16</u> measurements $\hat{\Pi}_\mu \otimes \hat{\Pi}_\nu$

↖ same projectors as Stokes.

$$I_{\mu\nu} = \mathcal{N} \, \mathrm{Tr}\{ \hat{\rho} \, (\hat{\Pi}_\mu \otimes \hat{\Pi}_\nu) \}.$$

$$= \frac{\mathcal{N}}{4} \sum_{\lambda k} \mathrm{Tr}\{ \hat{\sigma}_\lambda \hat{\Pi}_\mu \} \, \mathrm{Tr}\{ \hat{\sigma}_k \hat{\Pi}_\nu \} \, r_{\mu\nu}.$$

$$\left.\begin{aligned} \hat{\Pi}_0 &= \hat{\sigma}_0 \\ \hat{\Pi}_1 &= \tfrac{1}{2}(\hat{\sigma}_0 + \hat{\sigma}_1) \\ \hat{\Pi}_2 &= \tfrac{1}{2}(\hat{\sigma}_0 + \hat{\sigma}_2) \\ \hat{\Pi}_3 &= \tfrac{1}{2}(\hat{\sigma}_0 + \hat{\sigma}_3) \end{aligned}\right\} \Rightarrow \frac{1}{2} \mathrm{Tr}\{ \hat{\sigma}_\lambda \hat{\Pi}_\mu \} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 1/2 & 1/2 & 0 & 0 \\ 1/2 & 0 & 1/2 & 0 \\ 1/2 & 0 & 0 & 1/2 \end{pmatrix} = \Upsilon_{\lambda\mu}$$

$$\therefore \quad I_{\mu\nu} = \frac{\mathcal{N}}{4} \sum_{\lambda k} \Upsilon_{\lambda\mu} \Upsilon_{k\nu} \, r_{\mu\nu}.$$

Invert $\Upsilon_{\lambda\mu}$: "*Two-photon stokes parameter*".

$$S_{\mu\nu} = \sum_{\lambda k} \Upsilon^{-1}_{\mu\lambda} \Upsilon^{-1}_{\nu k} \, I_{\mu\nu}.$$

– again, no longer a direct observable.

$$\boxed{\hat{\rho} = \frac{1}{4} \sum_{\mu,\nu=0}^{3} \frac{S_{\mu\nu}}{S_{00}} \, \hat{\sigma}_\mu \otimes \hat{\sigma}_\nu}$$

– Easily generalized to n-qubits.

<u>Actually</u> it's a bit more complicated that this:

experimental noise → unphysical $\hat{\rho}$:

properties of $\hat{\rho}$:

i) $\mathrm{Tr}\{\hat{\rho}\} = 1$ (normalized)

ii) $\hat{\rho}^{+} = \hat{\rho}$ (Hermitian)

iii) Eigenvalues of $\hat{\rho}$ are all positive (positive-semi-definite)

(i), (ii) guarenteed by reconstruction, but not (iii)

Find "most likely" physical density matrix for given set of measurements

$$\mathcal{L}(\hat{\rho}) = \sum_{\mu,\nu=0}^{3} \frac{\left(I_{\mu\nu} - \mathrm{Tr}\{\hat{\rho}(\hat{\pi}_\mu \otimes \hat{\pi}_\nu)\}\right)^2}{\mathrm{Tr}\{\hat{\rho}(\hat{\pi}_\mu \otimes \hat{\pi}_\nu)\}}$$

data.

"guess" density matrix constrained to be "physical".

you know what projections were made!

$$\text{E.g.} \quad \hat{T}(t_1, t_2, \dots t_{16}) = \begin{pmatrix} t_1 & & & 0 \\ t_5 + it_6 & t_2 & & \\ t & t_7 + it_8 & t_3 & \\ \vdots & & \ddots & t_4 \end{pmatrix}$$

$$\text{then} \quad \hat{\rho}(t_1, t_2, \dots t_{16}) = \underbrace{\hat{T}^{+}\hat{T} \Big/ \mathrm{Tr}\{\hat{T}^{+}\hat{T}\}}_{\text{always satisfies the 3 criteria.}}$$

numerically maximise $\mathcal{L}$ over 16 parameters $t_1, \dots t_{16}$.

– Maximum likelihood estimation (or weight Least Squares)

– Hradil, Tan, D'Ariano.

Measures of Mixture

- Entropy $S = -\mathrm{Tr}\{\hat{\rho} \log_2 \hat{\rho}\}$.

$S = 0 \Rightarrow$ pure state

$S = \log_2 \underset{\uparrow}{N}$ ⇒ maximally mixed

dimension of Hilbert space

- "Linear Entropy" $S_L = [1 - \mathrm{Tr}\{\hat{\rho}^2\}.]$

$S = 0 \Rightarrow$ pure

$S_L = 1 \Rightarrow$ mixed

(no other meaning...)

Entropy → thermodynamics, Information theory, ...

Linear Entropy ⇒ No other meaning (but a bunch easier to calculate!)

Measures of Entanglement (2 qubits)

pure states:

$$|\psi\rangle = a|00\rangle + b|01\rangle + c|10\rangle + d|11\rangle$$

Concurrence $C = 2|ad - bc|$

closely related to Entanglement (defined by Carl):

$$\hat{\rho}_A = \mathrm{Tr}_B\{|\psi\rangle\langle\psi|\}$$

$$E = -\mathrm{Tr}\{\hat{\rho}_A \log_2 \hat{\rho}_A\}.$$
$$= h\left(\frac{1 + \sqrt{1 - C^2}}{2}\right)$$

$$h(x) = -x\log_2(x) - (1-x)\log_2(1-x)$$

Entanglement for Mixed States.

Make arbitrary decomposition of $\hat{\rho}$:

$$\hat{\rho} = \sum p_i |\psi_i\rangle\langle\psi_i|.$$

$|\psi_i\rangle$ are not orthogonal ⇒ not Eigenvalues.

"Average" Entanglement: $\sum_i p_i E(\psi_i)$.

can always decompose using Bell states: significant decompositio is the one for which this entanglement is a minimum.

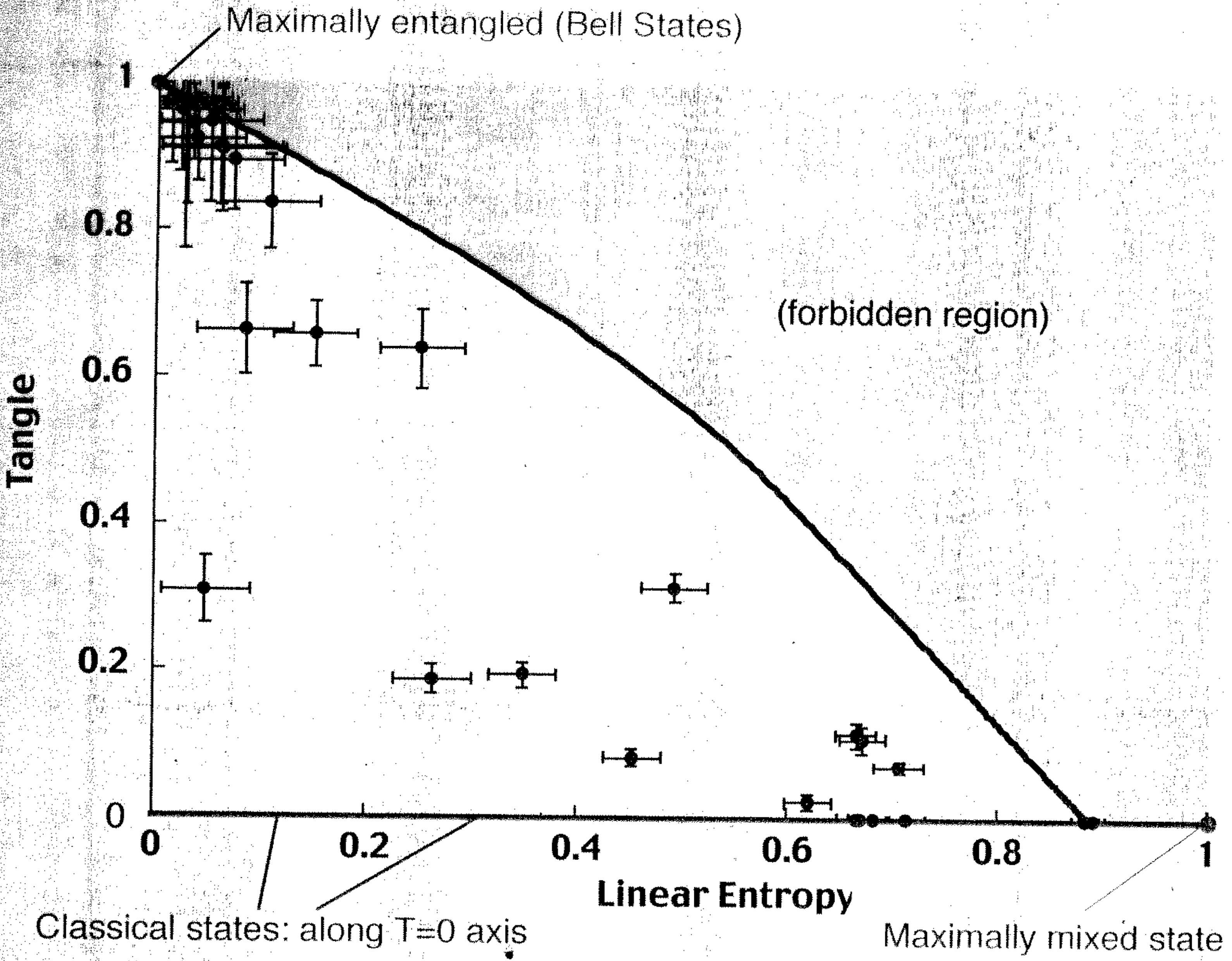
$$\mathcal{E}(\hat{\rho}) = \min_{\{\psi_i\}} \sum_i p_i E(\psi_i).$$

Woolers obtained a (complicated) explicit formula for this.

Maximum Entanglement for a given mixture:

$$\hat{\rho}_{MEMS} = \begin{pmatrix} g(c) & 0 & 0 & c/2 \\ 0 & 1-2g(c) & 0 & 0 \\ 0 & 0 & 0 & 0 \\ c/2 & 0 & 0 & g(c) \end{pmatrix}$$

→ "map" of Hilbert space, showing states we have created using two-photon down conversion.


Maximally entangled (Bell States)
Tangle
1
0.8
0.6
0.4
0.2
0
(forbidden region)
0
0.2
0.4
0.6
0.8
1
Linear Entropy
Classical states: along T=0 axis
Maximally mixed state

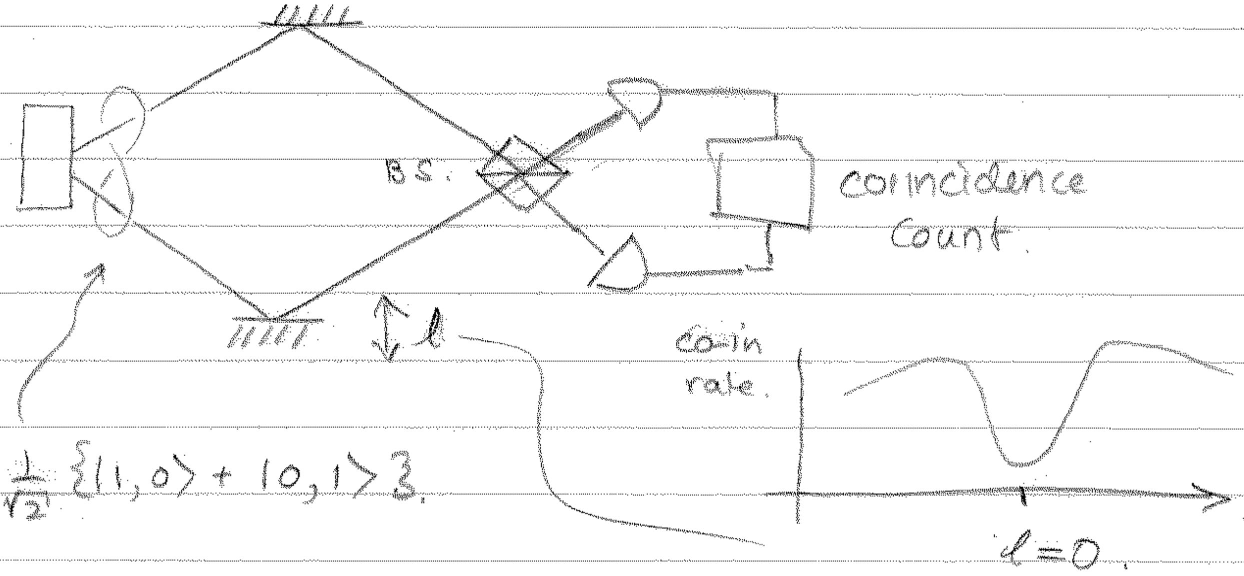
Two photon experiments for QIP.

- Good "factory" of entangled States (provided you don't mind post-selection)
- Single qubit operation
- quantum state measurement (efficiency 85% - 90% at best)
  - c.f. $\gtrsim 99\%$ efficiency for measurement of trapped ions.

- CANNOT do CNOT gates between photons.
  - weakness of $\chi^{(3)}$ non-linearity
  - attempts to get arround this (Franson)
  - Cannot do Bell state analysis needed for teleportation

Application of Entangled States:

- Tests of local realism (Bell inequalities)
- Hong-Ou-Mandel Interferometry
  - number state entanglement



state $\frac{1}{\sqrt{2}}\{|1,0\rangle + |0,1\rangle\}$.

- Quantum Teleportation / Dense Coding
- Quantum Cryptography: too complicated to go into in detail; but consider $|\Psi_-\rangle = \frac{1}{\sqrt{2}}(|HH\rangle + |VV\rangle)$
- Quantum Clock synchronization

suppose A & B share two qubits in a singlet state $|\Psi_-\rangle = \frac{1}{\sqrt{2}}(|01\rangle - |10\rangle)$ — ticking (non-degenerate) qubits.

Alice does a measurement of $\hat{\sigma}_x$ at time $t=0$.

$+1$: Bob's qubit in state $\frac{1}{\sqrt{2}}(|0\rangle - e^{i\omega_0 t}|1\rangle)$

-1: " " " " $\frac{1}{\sqrt{2}}(|0\rangle + e^{i\omega_0 t}|1\rangle)$

Bob measures σ_x at time t later with ensemble of qubits: $P_{++}(t) = \sin^2(\omega_0 t/2)$ — B can deduce t; BUT how to make $|\Psi_-\rangle$

Quantum Computing

Is entanglement necessary ?

Unitaries + measurement : can be done with pseudo-pure States just as with pure states. You are just recording the travel of a point on the surface of the Bloch sphere : it does not matter a priori how big the Bloch sphere is.

It can be small enough that entanglement is always precluded.

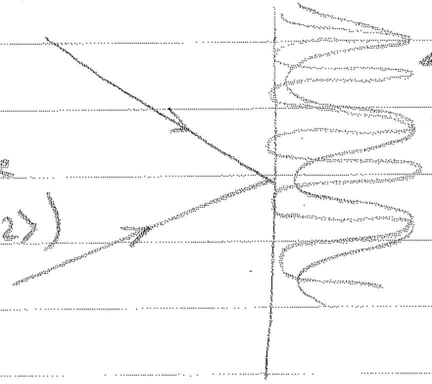
BUT : I would assert that, to be a quantum computer worthy of the name, it should do this things with a certain amount of efficiency ; in particular it is not going to be much use if you have a nice way to do a unitaries, but you have to repeat it all an exponentially large number of times because of poor measurement efficiency. - vide NMR (Linden & Popescu)

\- the criteria for good measurements & efficient unitaries may be enough to require entanglement.

(Parker & Plenio).

⑳

Quantum Lithography:



$$E(r)^4 :\rangle = \mathcal{E}_0^4 \left(\hat{a}_1 e^{i\underline{k}_1 . \underline{r}} + \hat{a}_2 e^{i\underline{k}_2 . \underline{r}} + \hat{a}_1^{+} e^{-i\underline{k}_1 . \underline{r}} + \hat{a}_2^{+} e^{-i\underline{k}_2 . \underline{r}}\right)^{4}$$

$: :$ –
normal ordering

$\overleftarrow{a^{+}} \quad \overrightarrow{a}$

"two photon pattern" $\langle : E(\underline{r})^4 : \rangle$

$$= 3|\mathcal{E}_0|^4 \{ \langle 2,0| \hat{a}_1^{+2} \hat{a}_2^{2} |0,2\rangle \exp[-i\, 2 \underline{\Delta k} . \underline{r}]$$
$$+ \underbrace{\langle 0,2| \hat{a}_2^{+2} \hat{a}_1^{2} |2,0\rangle}_{2} \exp[i\, 2 \underline{\Delta k} . \underline{r}] \}$$

(all other terms cancell)

$$= 12 |\mathcal{E}_0|^4 \cos(2 \underline{\Delta k} . \underline{r})$$