Two-atom excitation : with two-photon states of light

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Phys. Rev. A 88, 033822 (2013)

Question:

Is it possible to
increase or inhibit the probability
of a given process in light-matter interaction
by chosing appropriate quantum states of light ?

"Quantum coherent control" ?

Answer:

-no for linear processes
-perhaps for non-linear processes

2004 "M.A.S." paper:

LUME 93, NUMBER 9 PHYSICAL REVIEW LETTERS week endir
27 AUGUST

Inducing Disallowed Two-Atom Transitions with Temporally Entangled Photons

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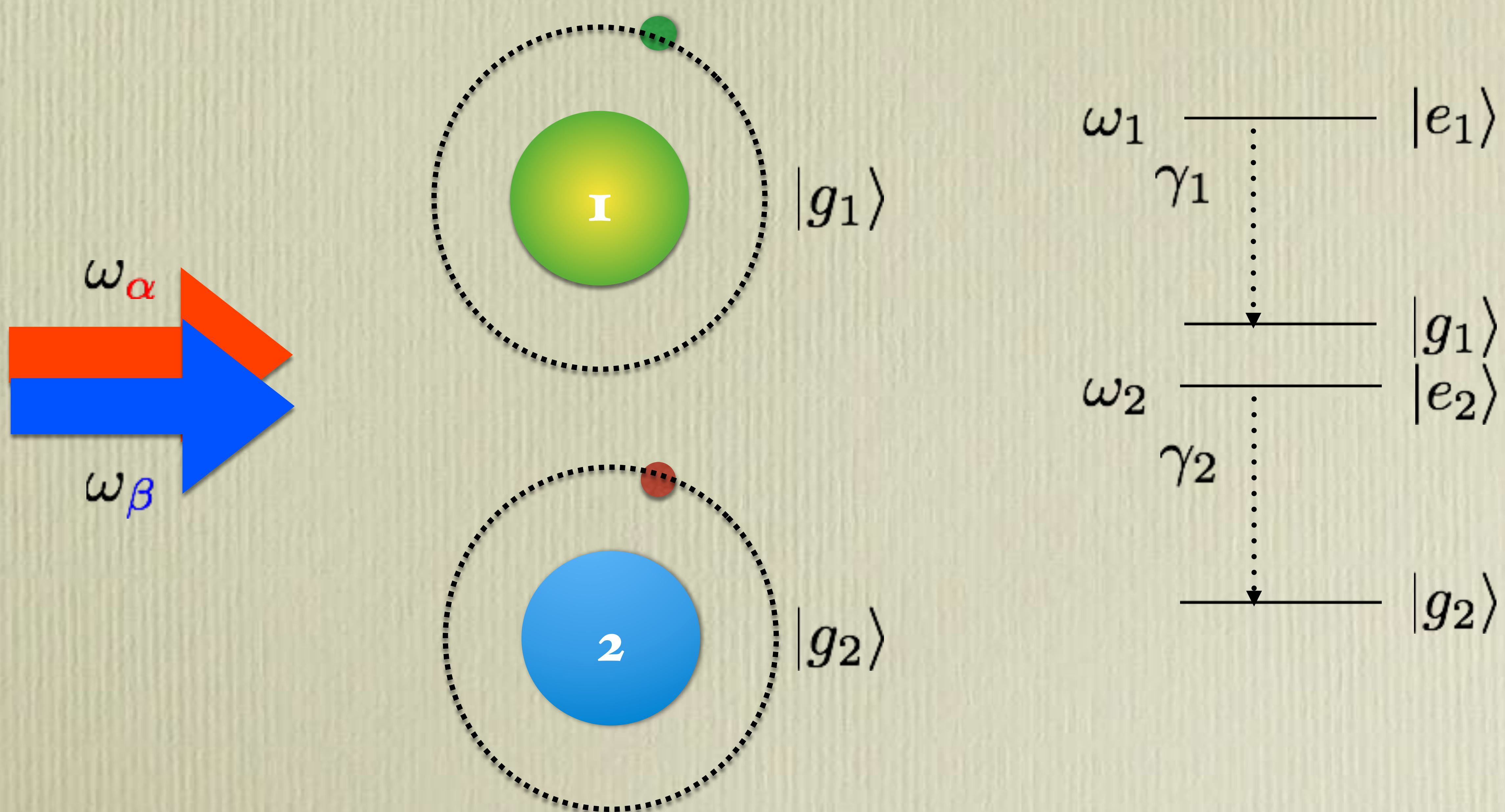
(Received 16 February 2004; published 25 August 2004)

Two uncoupled two-level atoms cannot be jointly excited by classical light under general circumstances, due to destructive interference of excitation pathways in two-photon absorption. However, with temporally entangled light, two-atom excitation is shown possible. Photons arising from three-level cascade decay are intrinsically ordered in time of emission. This field correlation induces a joint resonance in the two-atom excitation probability via suppression of one of the time-ordered excitation pathways. The relative gain in two-photon absorption increases with the time-frequency entanglement.

DOI: 10.1103/PhysRevLett.93.093002 PACS numbers: 32.80.–t, 42.50.Dv, 42.50.Hz

a striking quantum effect ??

the system:
two-photon excitation of 2 different atoms

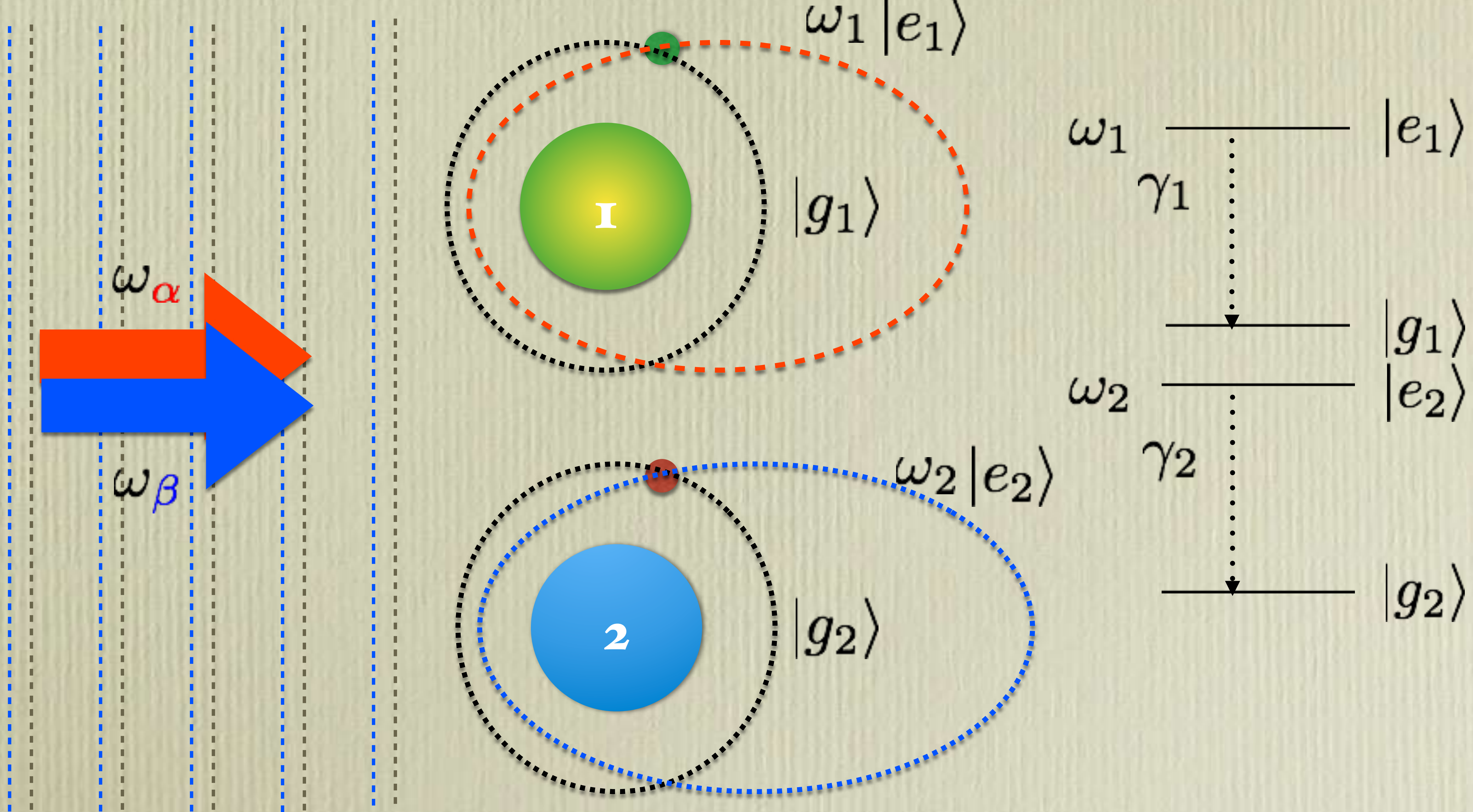


none of the photons is resonant with the atoms

the system:
two-photon excitation of 2 different atoms

the 2 photons are resonant with the 2 atoms

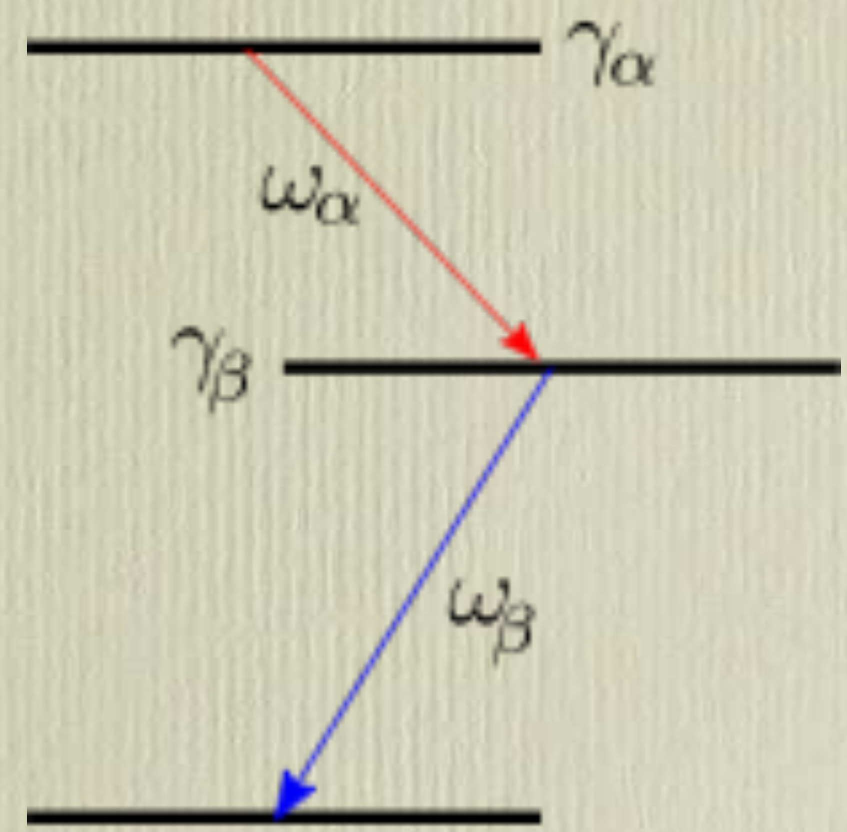
$$\boxed{\omega_\alpha + \omega_\beta \simeq \omega_1 + \omega_2}$$



none of the photons is resonant with the atoms

the quantum state considered by M.A.S.
to excite the two atoms:

a two-photon state
generated by atomic cascade

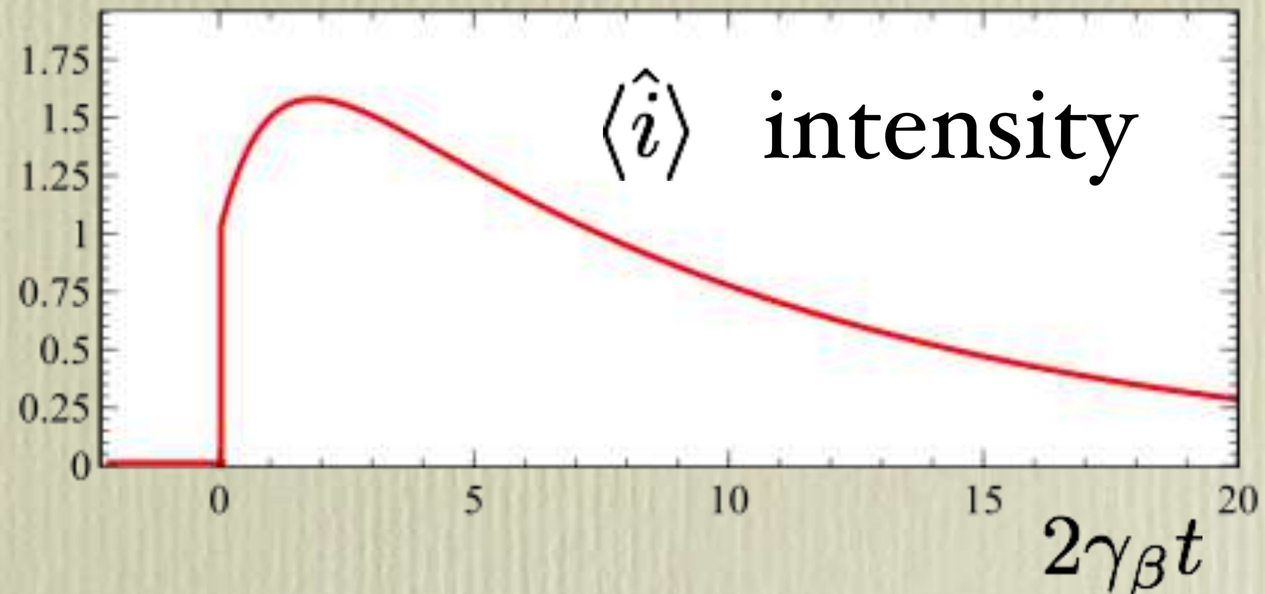


$$|cas\rangle = \sum_{kq} c_{kq}^{cas} |1:\omega_k; 1:\omega_q\rangle$$

$$c_{kq}^{cas} = \frac{g_\alpha(\omega_k) g_\beta(\omega_q)}{(\omega_{k\alpha} + \omega_{q\beta} + i\gamma_\alpha)(\omega_{q\beta} + i\gamma_\beta)}$$

M. O. Scully and M. S. Zubairy, *Quantum Optics*

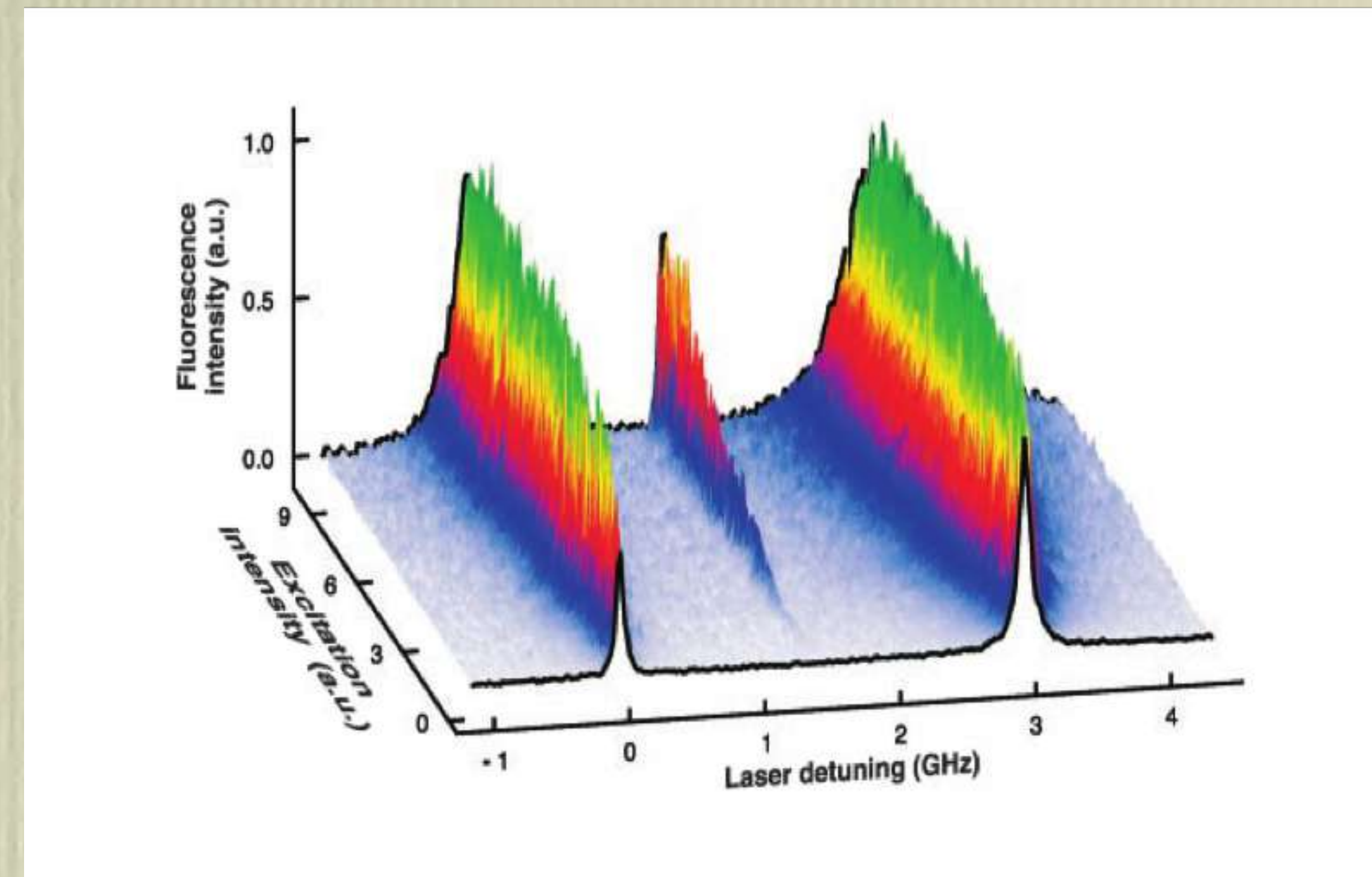
|cas> is an entangled state,



it describes a pulse of bi-photons starting at time t=0

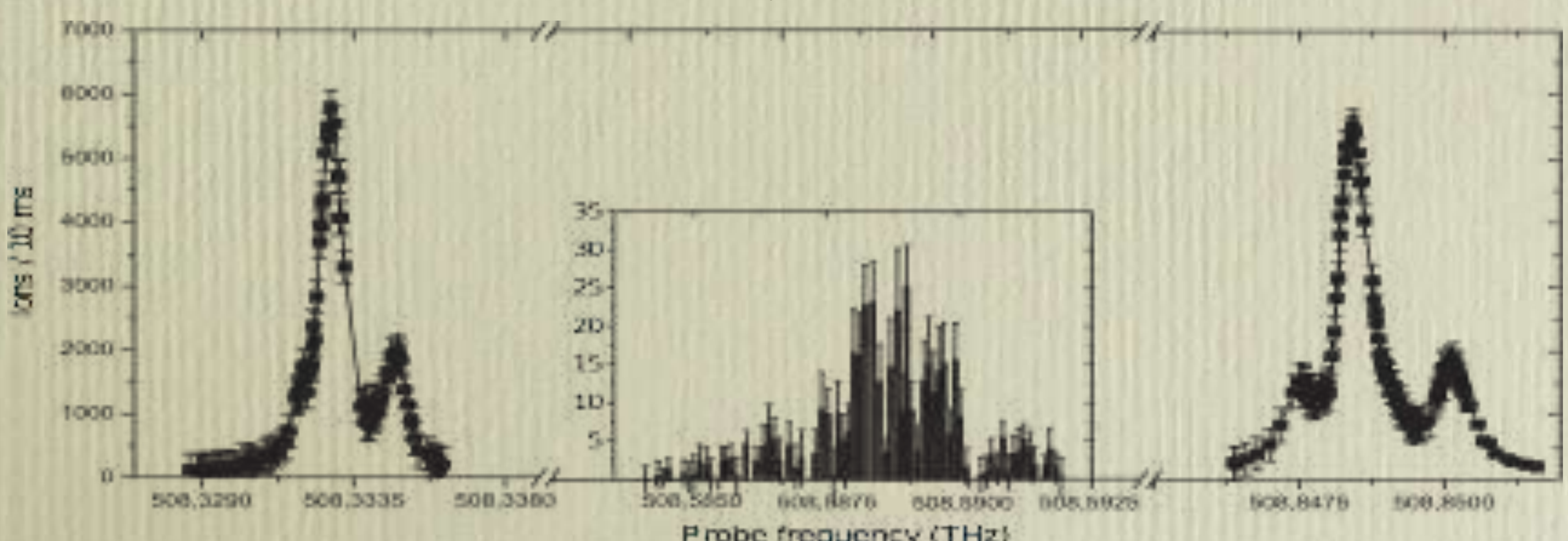
such a two-photon two-atom excitation is possible when the two atoms are related by some kind of interaction

J. R. Rios Leite, *et. al*, Chem. Phys. Lett. **73**, 71 (1980)



C. Hettich, *et. al*, Science. **298**, 385 (2002)

E. Pedrozo-Peñafiel, *et. al*,, Phys. Rev. Lett. **108**, 253004 (2012)



In some situations,
can entanglement replace
a real physical interaction
?

a new important property of entanglement !

Complete quantum calculation of 2-photon 2-atom excitation

(lowest order perturbation theory, long lifetimes of both atoms)

stationary excitation probability by two-photon state described by density matrix ρ_o:

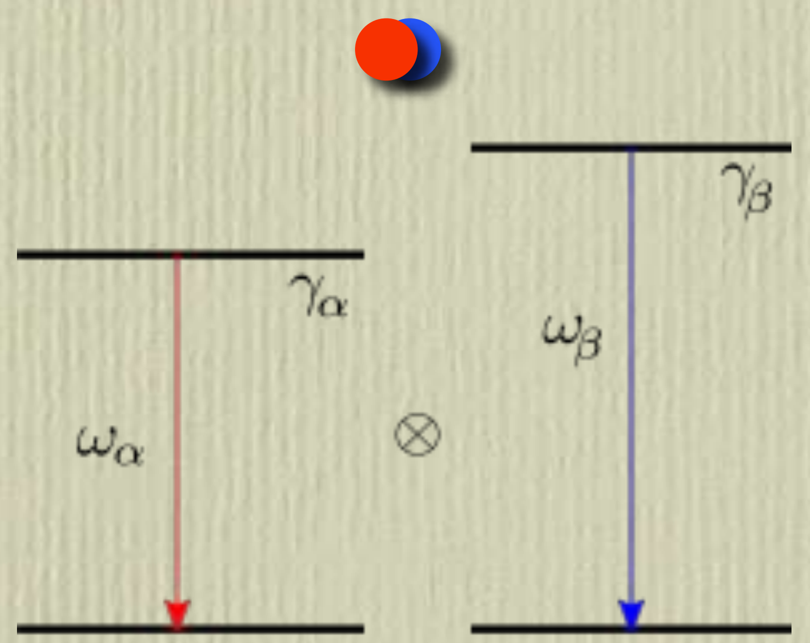
$$P(t) \approx \sum_{jkmn} \mathcal{A}^*_{jk} \mathcal{A}_{mn} \mathrm{Tr}(a^\dagger_j a^\dagger_k a_m a_n \rho_0)$$

$$\mathcal{A}_{mn} = f_1(\omega_m) f_2(\omega_n) \frac{1 - e^{i(\omega_1 - \omega_m)t}}{\omega_m - \omega_1} \frac{1 - e^{i(\omega_2 - \omega_n)t}}{\omega_n - \omega_2}$$

product of single photon response functions

Uncorrelated bi-photon source:

Uncorrelated spontaneous emission photons emitted by two independent atoms



$$|\mathrm{I}\otimes\mathrm{I}\rangle = \sum_{kq} c^{11}_{kq}|1:\omega_k;1:\omega_q\rangle$$

$$c^{11}_{kq} = \frac{g_\alpha(\omega_k)g_\beta(\omega_q)}{(\omega_k-\omega_\alpha+i\gamma_\alpha)(\omega_q-\omega_\beta+i\gamma_\beta)}$$

factorized state

two-atom transition probability for factorized state:

at 2-photon 2-atom resonance:

$\omega_1 + \omega_2 = \omega_\alpha + \omega_\beta$ but large single photon detunings

$$P^{11}_{2P2A} = \frac{P_0 \gamma_\alpha \gamma_\beta}{\Delta^4} = P^{11}_{DR} \frac{\gamma_\alpha^2 \gamma_\beta^2}{\Delta^4}$$

transition probability
in the doubly resonant case

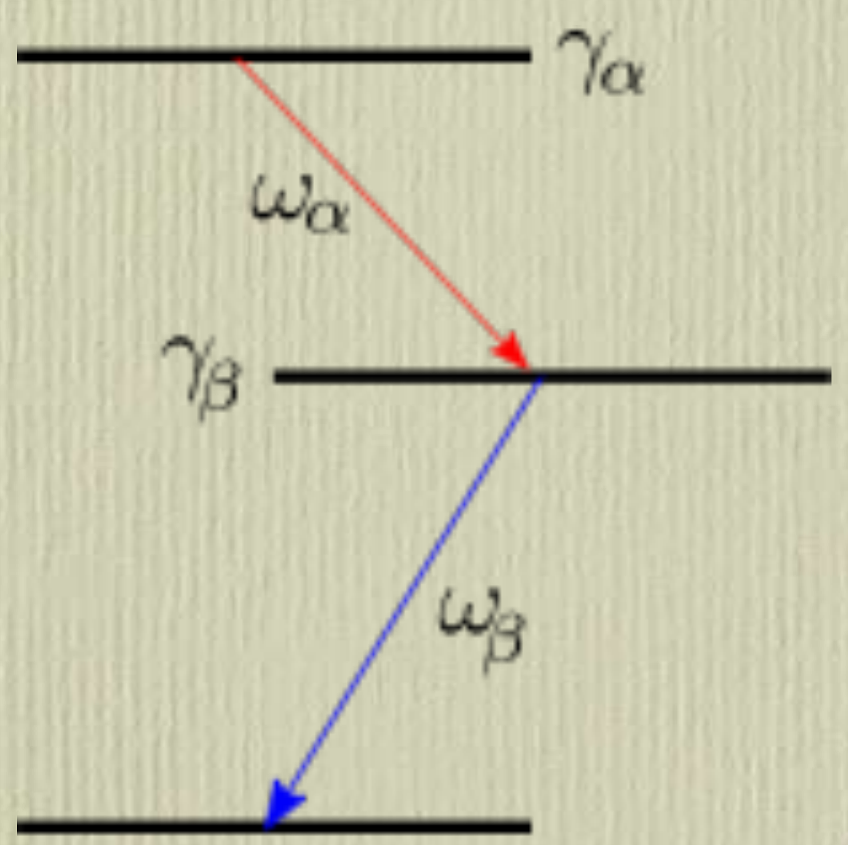
$$\Delta = \omega_\alpha - \omega_1 = \omega_2 - \omega_\beta$$

single photon detuning

no two-photon resonance

Entangled bi-photon source:

M.A.S entangled cascade state



Probability: $\simeq P_{DR}^{11}\frac{\gamma_\beta^2}{\Delta^2}$

resonant character
around two-photon energy match

Enhancement with respect
to two independent photons:

$$\frac{P_{|\mathrm{cas}\rangle}}{P^{|\mathrm{cas}\rangle}_{\mathrm{dep,fac}}} \simeq \frac{\Delta^2}{\gamma_\alpha(\gamma_\alpha+\gamma_\beta)} \gg 1.$$

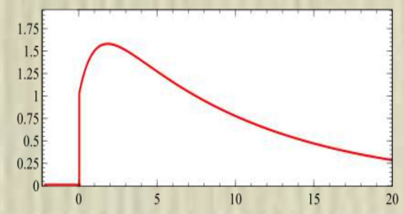
M.A.S. result is right
enhancement factor can be very large

Is the M.A.S entangled state
the only quantum state
likely to enhance
the two-photon two-atom transition probability
??

From any pure two-photon state

t=0 $$|\Psi\rangle = \sum_{kq} c_{kq} |1:\omega_k; 1:\omega_q\rangle$$

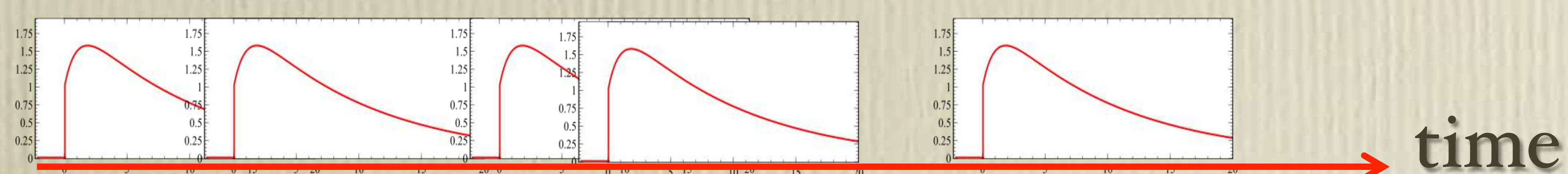
which describes a pulse of biphotons



a) one can build a mixed state
by randomizing the starting time of the cascade

$$\Lambda = \sum_{kq} |c_{kq}|^2 |1:\omega_k; 1:\omega_q\rangle\langle 1:\omega_k; 1:\omega_q|$$

- it is a separable state, with some degree of correlation
- it describes c.w. light



b) one can build a factorized state
with same energy and same frequency spectrum

One finds that the two-atom excitation probability
is roughly the same

- for the M.A.S. entangled cascade pure state

-for the corresponding separable mixed state

provided that one interrupts the interaction
when the energy carried by c.w. light
is equal to the cascade pulsed state energy

Both have a resonant character $\frac{1}{\delta^2 + \gamma_\alpha^2}$