

Title: TBA

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Abstract:

Sharpening accepted thermodynamic wisdom via quantum control: or cooling to an internal temperature of zero by external coherent control fields without spontaneous emission

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(Received 15 February 2002; revision received 10 May 2002)

Abstract. Cooling of internal atomic and molecular states via optical pumping and laser cooling of the atomic velocity distribution, rely on spontaneous emission. The outstanding success of such examples, taken together with general arguments, has led to the widely held notion that radiative cooling requires spontaneous emission. We here show by specific examples and direct calculation, based primarily on breaking emission–absorption symmetry as in lasing without inversion, that cooling of internal states by external coherent control fields is possible. We also show that such coherent schemes allow us to practically reach absolute zero in a finite number of steps, in contrast to some statements of the third law of thermodynamics.



“About your cat, Mr. Schrödinger—I have good news and bad news.”



Quantum Coherence Can Improve Quantum Heat Engine (QHE) Efficiency

I. Quantum Thermodynamics

II. Photo-Carnot QHE

III. The Laser as a QHE

1. $\frac{\hbar\nu_{laser}}{\hbar\nu_{pump}} = 1 - \frac{T_c}{T_h}$ From Detailed Balance

2. Quantum Coherence Can Improve Laser QHE Quantum Efficiency by
Breaking Detailed Balance

IV. The Photocell as a QHE

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V. Noise Induced Quantum Coherence Can Enhance:

1). Laser QHE Power

2). Photocell QHE Power

3). Efficiency of Photosynthesis



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Heat Engines

- **Classical Heat Engines** produce useful work by extracting energy from a high temperature energy source and rejecting entropy to a low temperature entropy sink.
- **A Photo-Carnot Engine** is a Carnot cycle engine in which photons are the working fluid and the piston is driven by radiation pressure. Quantum coherence allows us to achieve thermodynamic efficiency beyond the Carnot limit without violating the second law.
- **Laser and Photocell Quantum Heat Engines** are driven by thermal radiation and governed by the laws of quantum thermodynamics.



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Quantum Thermo I

Planck studies **Entropy** of Light to Arrive at the **Quantum***

Wein Entropy

$$\frac{\partial^2 S}{\partial \bar{\epsilon}^2} = -\frac{1}{b \omega \bar{\epsilon}}$$

where $\bar{\epsilon}$ = average energy of
oscillator with
frequency ω

Planck Entropy 1900

$$\frac{\partial^2 S}{\partial \bar{\epsilon}^2} = -\frac{k_B}{\bar{\epsilon}(\hbar \omega - \bar{\epsilon})}$$



$$\bar{\epsilon} = \frac{\hbar \omega}{e^{\hbar \omega / k_B T} - 1}$$

*Planck Photon Statistics and Bose Einstein condensation, Progress in Optics 2007



Quantum Thermo II

Einstein studies **Entropy** of Light to Arrive at the **Photon***

Fluctuations

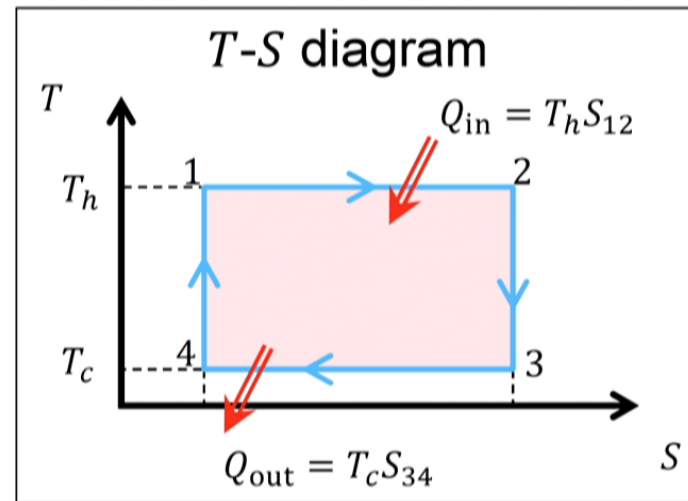
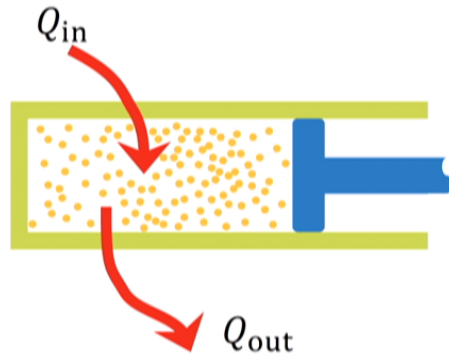
$$\frac{\langle (\Delta E)^2 \rangle}{(\hbar \omega)^2} = \underbrace{\bar{n}^2}_{\text{wave}} + \underbrace{\bar{n}}_{\text{particle}}$$

Wave Particle Duality 1905

*Progress in Optics 2007



Carnot Efficiency

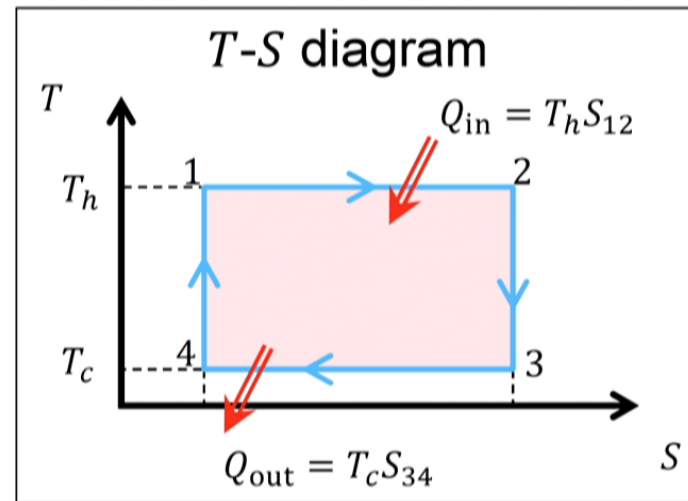
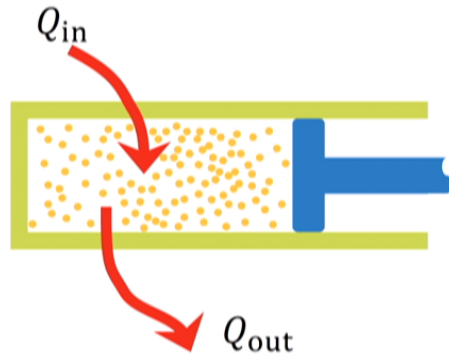


thermal efficiency

$$\eta = \frac{Q_{in} - Q_{out}}{Q_{in}} = \frac{T_h S_{12} - T_c S_{34}}{T_h S_{12}} = 1 - \frac{T_c}{T_h}$$



Carnot Efficiency



thermal efficiency

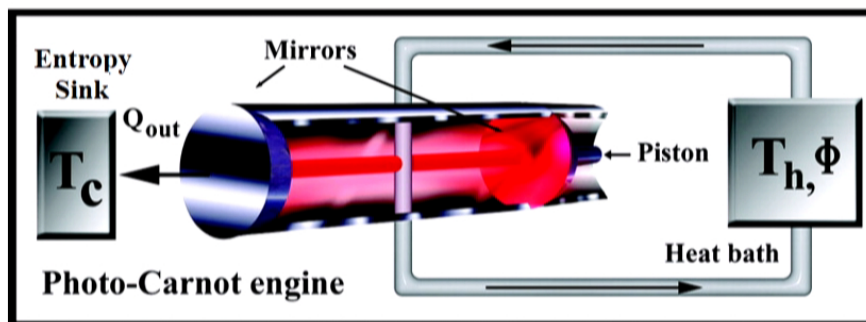
$$\eta = \frac{Q_{in} - Q_{out}}{Q_{in}} = \frac{T_h S_{12} - T_c S_{34}}{T_h S_{12}} = 1 - \frac{T_c}{T_h}$$



Photo-Carnot Engine

- Working Fluid(radiation) heated by two-level atoms

$$PV = \hbar\Omega\bar{n}$$



Regular

$$\eta = 1 - \frac{T_c}{T_h}$$



Extracting Work from a Single Heat Bath via Vanishing Quantum Coherence

M. Scully, M. Zubairy, G. Agarwal, and H. Walther

Science **299**, 862 (2003);
DOI: 10.1126/science.1078955



One atom

$$PV = kT$$

One mode

$$PV = \hbar \Omega \bar{n}$$

$$\frac{1}{e^{\frac{\hbar \Omega}{kT}} - 1} \rightarrow \frac{kT}{\hbar \Omega}$$

Single Mode = Single Atom

$$PV = kT \quad (\text{One atom})$$

$$PV = \hbar\Omega\bar{n} \quad \bar{n} = \frac{kT}{\hbar\Omega}$$

$$= kT \quad (\text{One mode})$$

$$\eta_c = 1 - \frac{T_c}{T_h}$$



Rate equation for photon number (I)

- With two-level atoms field,

$$\dot{\bar{n}} = \alpha[\rho_{aa}(\bar{n} + 1) - \rho_{bb}\bar{n}]$$

- The steady-state solution is

$$\bar{n} = \frac{1}{\frac{\rho_{bb}}{\rho_{aa}} - 1} = \frac{1}{\exp(\hbar\Omega/kT_h) - 1} \xrightarrow[\hbar\Omega/kT_h \ll 1]{\text{high temperature}} \frac{kT_h}{\hbar\Omega}$$



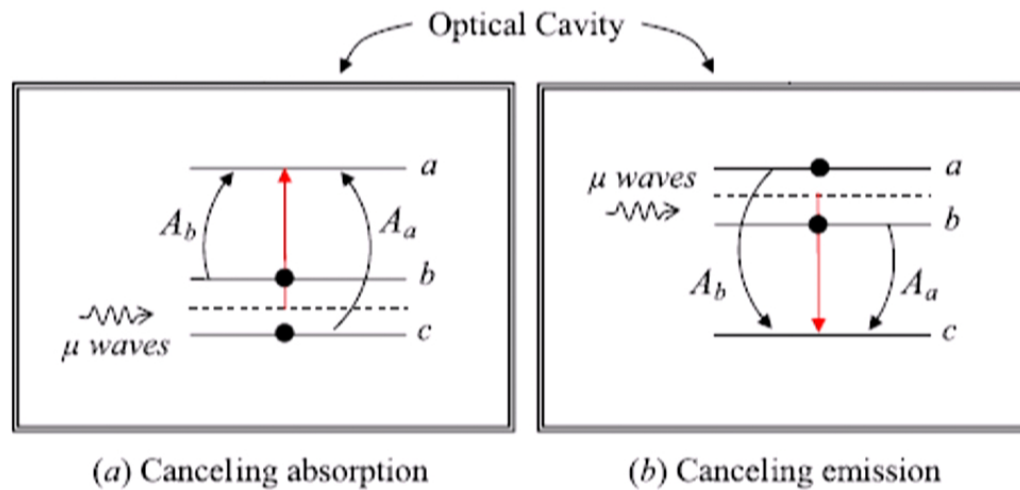
— a p_{aa}

— b p_{bb}

— a ρ_{aa}

— b ρ_{bb}

Lasing Without Inversion



- (a) Use of quantum coherence in ground state b, c to cancel absorption
- (b) the use quantum coherence in the excited state a, b to cancel emission

$$\dot{\bar{n}}_{laser} = \alpha(\bar{n} + 1)(|A_a|^2 + |B_b|^2) - \alpha\bar{n}|A_a + B_b|^2$$

Laser Oscillation without Population Inversion in a Sodium Atomic Beam

G. G. Padmabandu,^{1,2,*} George R. Welch,^{1,2} Ivan N. Shubin,¹ Edward S. Fry,^{1,2,†} Dmitri E. Nikonov,^{1,2,‡}
Mikhail D. Lukin,^{1,2} and Marlan O. Scully^{1,2,3}

¹Texas Laser Laboratory, Houston Advanced Research Center, The Woodlands, Texas 77381

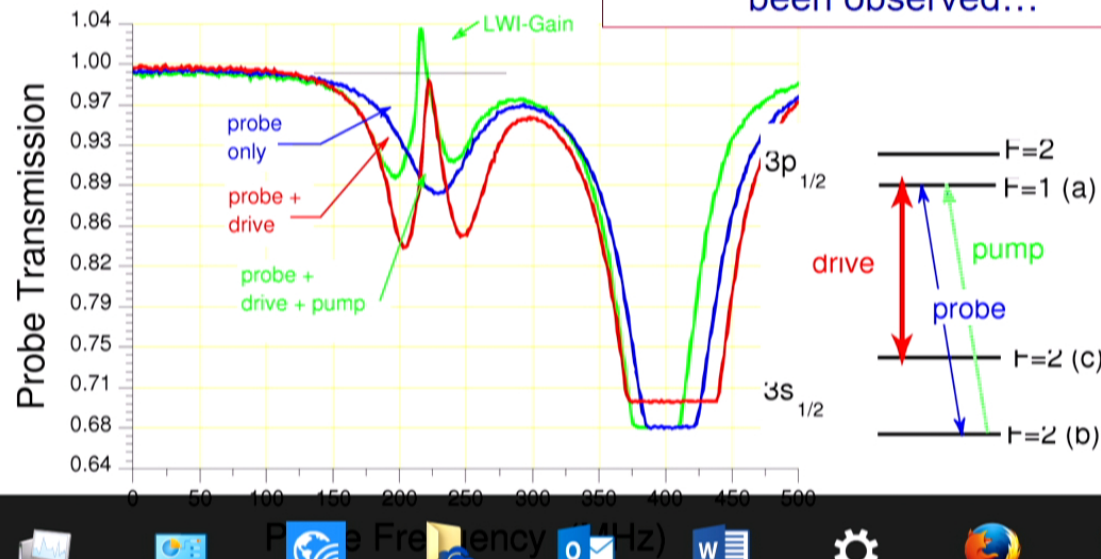
²Department of Physics, Texas A&M University, College Station, Texas 77843-4242

³Max-Planck-Institut für Quantenoptik, D-85748 Garching, Germany

(Received 29 August 1995)

Continuous wave (cw) amplification and laser oscillation without population inversion have been observed for the first time in a Λ scheme within the sodium D_1 line. This is also the first demonstration in which the lasing medium was an atomic beam; this is the first demonstration of the physics, lays a foundation for extensions into the ultraviolet, and the atomic structure were critical to the choice of experimental apparatus. Theoretical matrix calculations and clearly show there was no population inversion.

“Continuous wave (cw) amplification and laser oscillation without population inversion have been observed...”

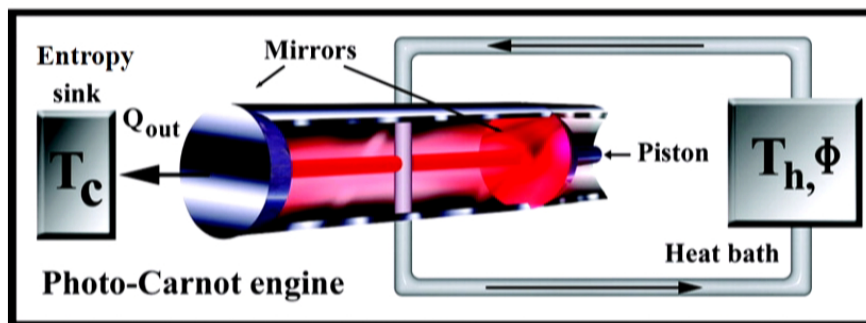


10% — a ρ_{aa}

45% — b ρ_{bb}
45% — c

Photo-Carnot Engine

- Working Fluid heated by phaseonium



Phaseonium

$$\eta_\phi = \eta - \xi \cos \phi$$

The diagram shows three energy levels labeled a, b, and c. Level a is the highest, level b is in the middle, and level c is the lowest. A red wavy arrow labeled Ω points from level c to level a.



Rate equation for photon number (II)

- With phased three level atoms(phaseonium) field,
$$\dot{\bar{n}}_{\phi} = \alpha [2\rho_{aa}(\bar{n}_{\phi} + 1) - (\rho_{bb} + \rho_{cc} + \rho_{bc} + \rho_{cb})\bar{n}_{\phi}]$$
- The steady-state solution is

$$\bar{n}_{\phi} \xrightarrow[\hbar\Omega/kT_h \ll 1]{\text{high temperature}} \frac{kT_h}{\hbar\Omega} \left(1 - \bar{n} \frac{|\rho_{bc}|}{\rho_{aa}} \cos \phi \right) \equiv \frac{kT_{\phi}}{\hbar\Omega}$$

T_{ϕ} : effective radiation temperature



10% —•— a

ρ_{aa}

45% —•— b

45% —•— c

ρ_{bb}

$$\vec{\rho}(\rho_{ab} + cc) = \vec{p}$$

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10% —•— a

p_{aa}

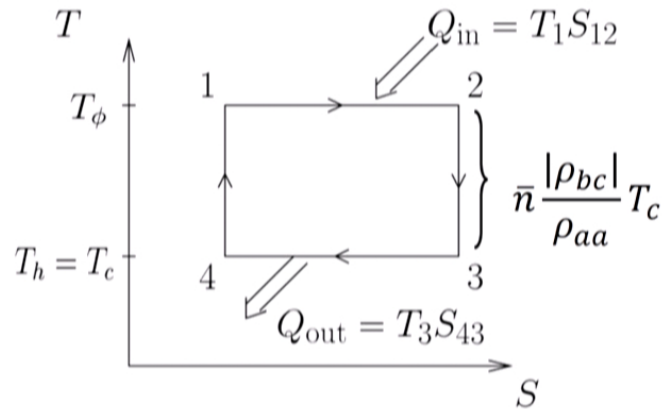
45% —•— b

45% —•— c

p_{bb}

$$\vec{p}(p_{bc} + cc) = \vec{p}$$

Efficiency of a Quantum Carnot Engine



Temperature-entropy diagram for Carnot cycle engine.

In the present QHE, Q_{in} is provided by the hot atoms.

When $T_h = T_c$, the photo-Carnot engine can still produce useful work if the

coherent three-level heat bath atoms are

"biased" such that $\phi \neq 0$

$$\eta = \frac{Q_{in} - Q_{out}}{Q_{in}}$$

$$\begin{aligned} \eta_\phi &= \frac{T_\phi S_{12} + T_c S_{43}}{T_\phi S_{12}} \\ &= \eta - \frac{T_c}{T_h} \bar{n} \frac{|\rho_{bc}|}{\rho_{aa}} \cos \phi \end{aligned}$$



10% — a

p_{aa}

45% — b
45% — c

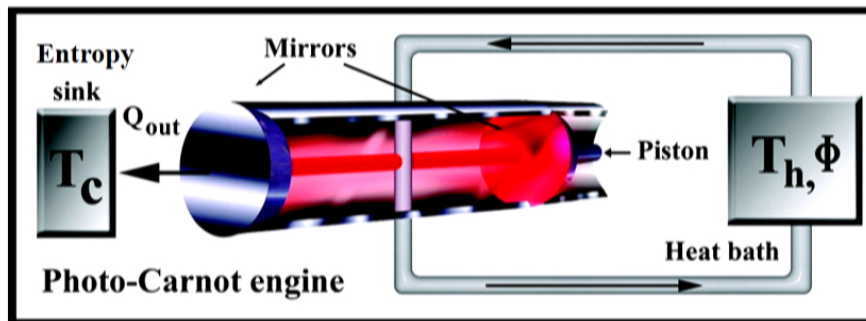
p_{bb}

$$\frac{1}{8}(p_{bc} + cc) = \bar{p}$$

$$\begin{pmatrix} p_{aa} & 0 & 0 \\ 0 & p_{bb} & p_{bc} \\ 0 & p_{cb} & p_{cc} \end{pmatrix}$$

Photo-Carnot Engine

- Working Fluid heated by phaseonium



Phaseonium

$$\eta_\phi = \eta - \xi \cos \phi$$

The diagram shows three energy levels labeled a, b, and c. A red wavy arrow labeled Ω indicates a transition from level b to level a.



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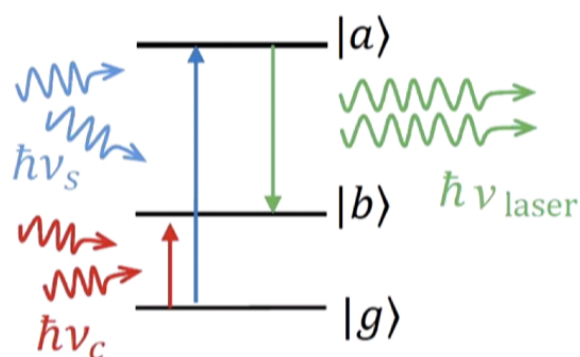
Laser Quantum Heat Engine (QHE)

PRL 2, 262 (1959) PHYSICAL REVIEW LETTERS

MARCH 15, 1959

THREE-LEVEL MASERS AS HEAT ENGINES*

H. E. D. Scovil and E. O. Schulz-DuBois
Bell Telephone Laboratories,
Murray Hill, New Jersey
(Received January 16, 1959)



Boltzmann distribution

$$\frac{n_b}{n_a} = \frac{n_b}{n_g} \cdot \frac{n_g}{n_a} = e^{-\hbar\nu_c/kT_c} \cdot e^{\hbar\nu_s/kT_h}$$

At threshold ($n_b = n_a$) efficiency of maser QHE

$$\frac{\hbar\nu_\ell}{\hbar\nu_s} = 1 - \frac{T_c}{T_s}$$



$$\bar{n}_s = \frac{1}{e^{\frac{\hbar\omega_{ab}}{kT_s}} - 1}$$

T_s
 \sim
 \sim

a
 T_h
 b
 c

$$\bar{n}_s = \frac{1}{e^{\frac{\hbar \omega_{ab}}{k T_s}} - 1}$$

T_s
 T_h
 a
 b
 g

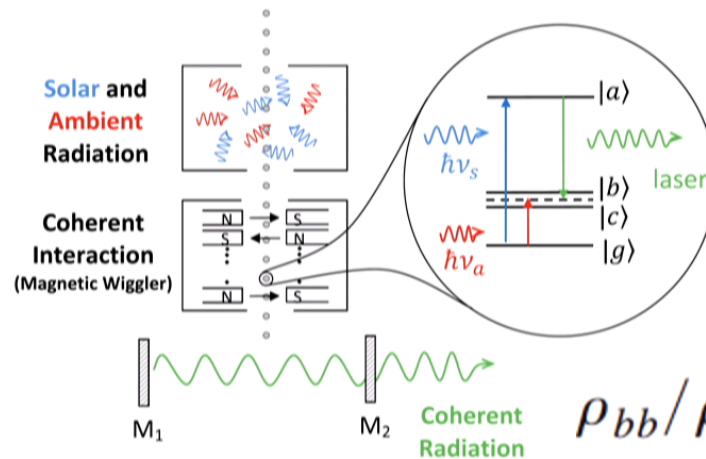
$$\omega_{ab} = \frac{E_a - E_b}{\hbar} = \nu_{hs}$$

$$\bar{n}_s = \frac{1}{e^{\frac{\hbar \omega_{ab}}{k T_s}} - 1}$$

T_s
 T_h
 a
 b
 g

$$\omega_{ab} = \frac{E_a - E_b}{\hbar} = \nu_{hs}$$

$$\frac{\hbar \nu_c}{k T_c} = \frac{\hbar \nu_s}{k T_h}$$



Marlan O. Scully
Texas A&M University
College Station
Texas 77843, USA and
Princeton University
Princeton, New Jersey 08544, USA

$$\rho_{bb}/\rho_{aa} = \exp[-\epsilon_b/kT_c - \epsilon_a/kT_h]$$

$$\dot{\mathcal{E}} = \kappa(2\rho_{aa} - \rho_{bb} - \rho_{cc} - \rho_{bc} - \rho_{cb})\mathcal{E}$$

At threshold

$$\frac{\hbar\nu_\ell}{\hbar\nu_s} = \eta_{\text{Carnot}} + \delta\eta$$

$$\delta\eta = kT_c|\rho_{bc}|/\hbar\nu_s\rho_{aa}$$



22



Summary

- Efficiency from QHE can exceed the classical Carnot Efficiency by using phased three-level atoms.
- The Photo-Carnot QHE can produce work from a single thermal bath.
- Efficiency of Laser QHEs can be increased by quantum coherence.



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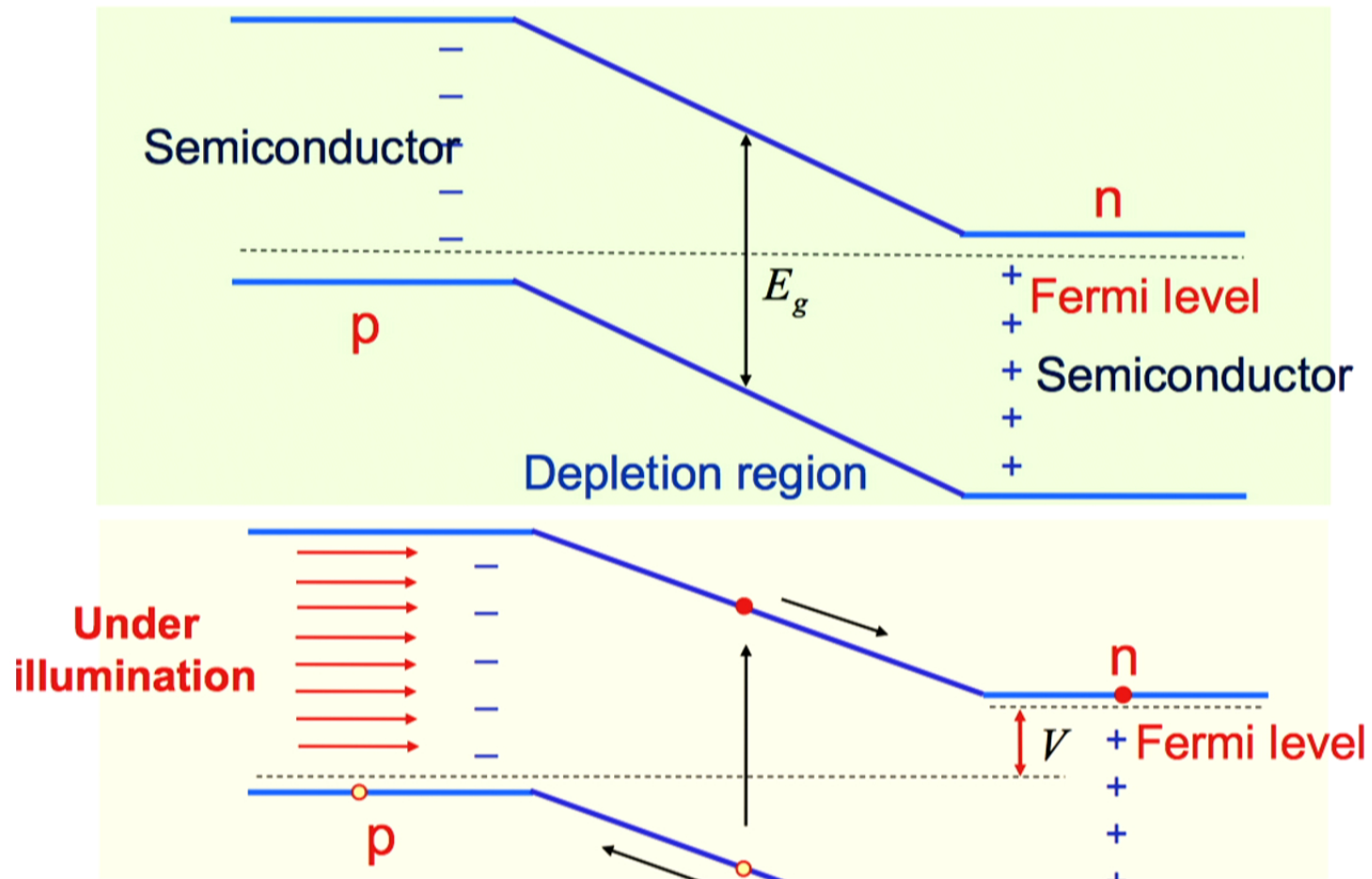
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2). Photocell QHE Power

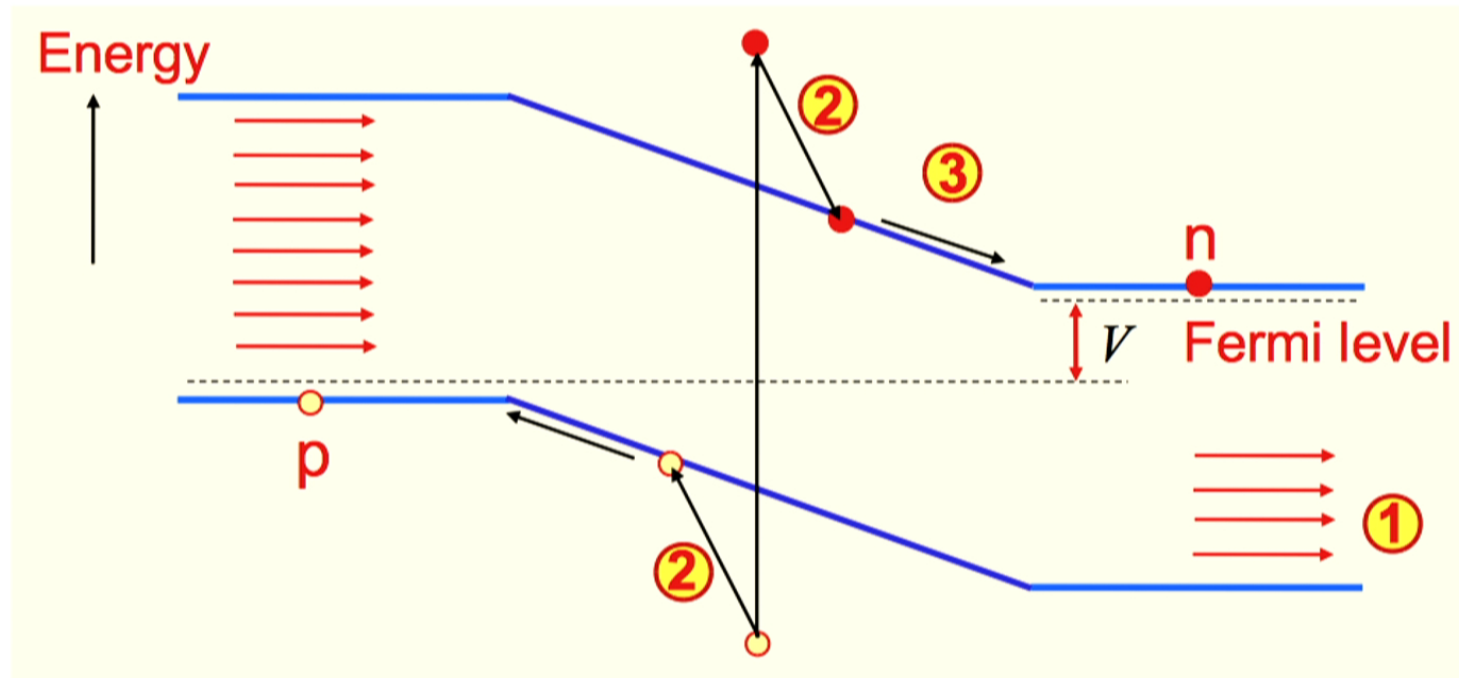
3). Efficiency of Photosynthesis



pn-junction solar cell



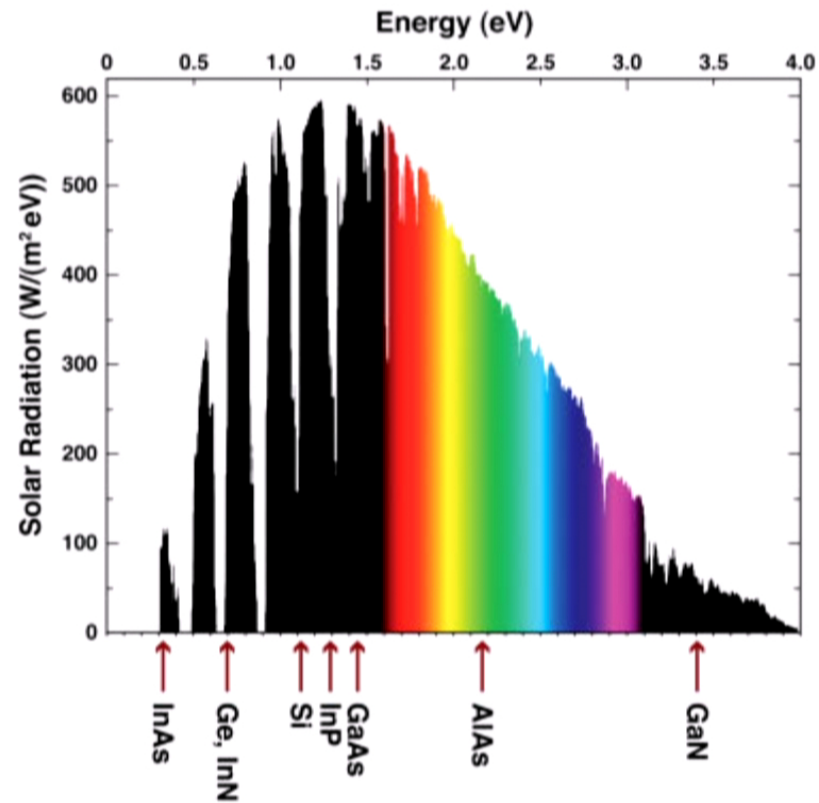
Solar cell losses



1. Photons with energy less than E_g are not absorbed
2. Thermal relaxation, energy is lost to phonons

3. Losses due to finite temperature of the cell (thermodynamic)



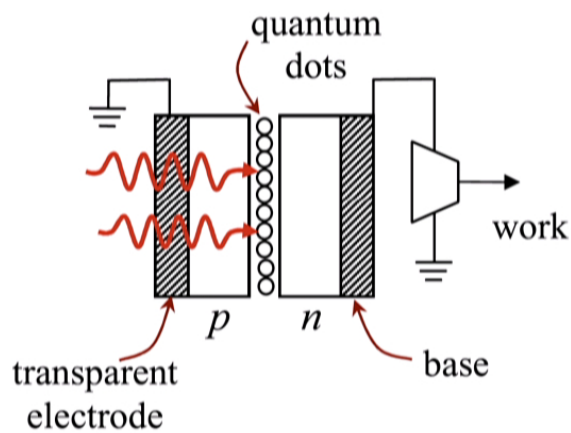


Solar spectrum with the bandgaps of semiconductors

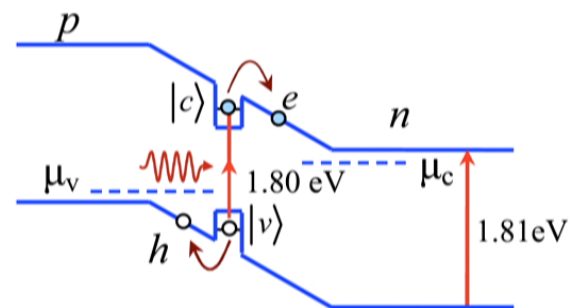


Quantum Dot Photo/Solar Cell

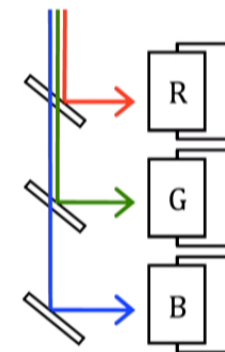
Solar cell with array of quantum dots



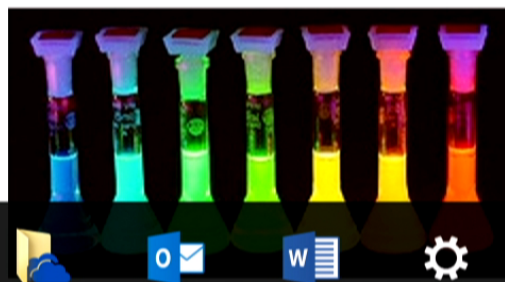
Electron-hole separation



Dividing photon flux onto monochromatic components



Texas A&M
University



PRINCETON
UNIVERSITY



Solar Cells and Detailed Balance

PRL **104**, 207701 (2010)

PHYSICAL REVIEW LETTERS

week ending
21 MAY 2010



Quantum Photocell: Using Quantum Coherence to Reduce Radiative Recombination and Increase Efficiency

Marlan O. Scully

Texas A&M University, College Station, Texas 77843, USA

Princeton University, Princeton, New Jersey 08544, USA

(Received 18 November 2009; published 21 May 2010)

The fundamental limit to photovoltaic efficiency is widely thought to be radiative recombination which balances radiative absorption. We here show that it is possible to break detailed balance via quantum coherence, as in the case of lasing without inversion and the photo-Carnot quantum heat engine. This yields, in principle, a quantum limit to photovoltaic operation which can exceed the classical one. The present work is in complete accord with the laws of thermodynamics.

P. Würfel, *Chimia* **61**, 770 (2007)

“That leaves radiative recombination as the major [energy loss] process. Can this be avoided? The answer is no. If a radiative upward transition to generate the excitation is allowed, its reversal, the radiative downward transition must be allowed as well.”



Solar cell with detailed balance

Population on levels c and v at temperature T_a

$$\frac{N_v}{N_c} = \exp[\epsilon_c - \epsilon_v - (\mu_c - \mu_v)]/kT_a$$

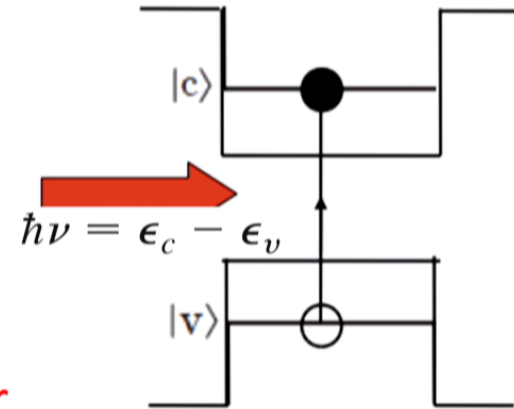
$$eV = \mu_c - \mu_v \quad \text{Cell voltage}$$

Average photon number

$$\dot{\bar{n}} = -R \left[\frac{1}{e^{\hbar\nu/kT_s} - 1} - \frac{1}{e^{(\hbar\nu - eV)/kT_a} - 1} \right],$$

Thermodynamic efficiency

$$eV = \hbar\nu \left(1 - \frac{T_a}{T_s} \right)$$



Solar cell with external coherence

External drive $\hbar\nu_0 = \frac{1}{2}(\epsilon_{c1} - \epsilon_{c2})$

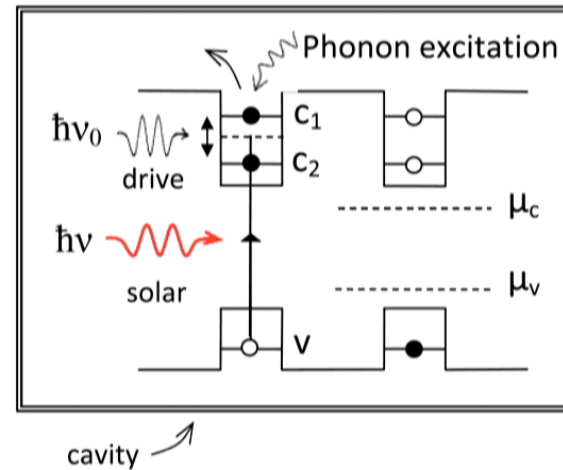
Resonant interaction $\Omega = \omega_1 - \nu$

Average photon number

2-level

3-level

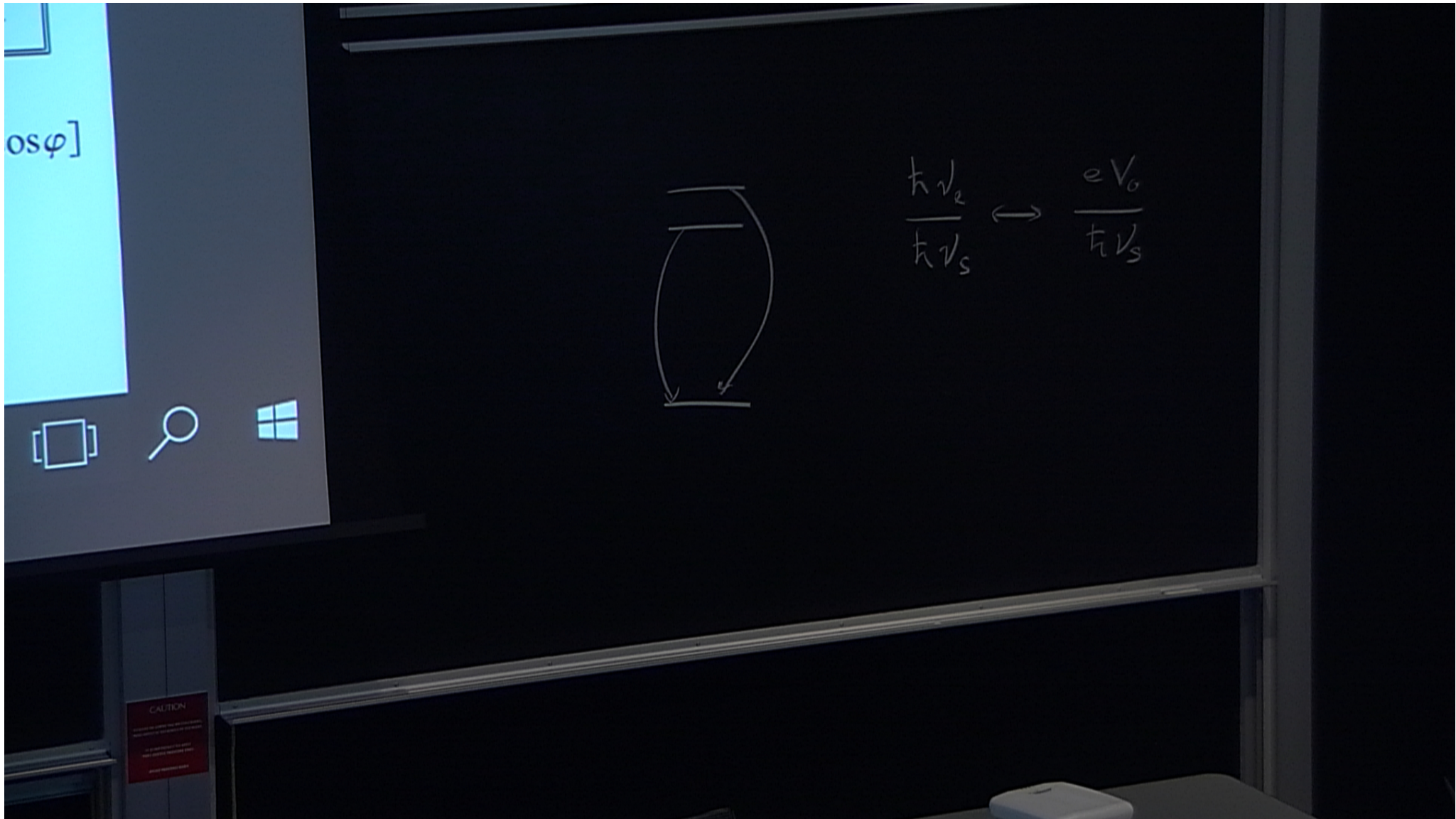
$$\frac{1}{e^{(\hbar\nu - eV)/kT_a} - 1} \rightarrow \frac{1}{e^x - 1} \quad e^x = 2\rho_{v,v}/[\rho_{1,1} + \rho_{2,2} + (\rho_{11} - \rho_{22})\cos\varphi]$$



Thermodynamic efficiency $\varphi = \pi$

$$eV = \hbar\nu \left(1 - \frac{T_a}{T_s} \right) + \hbar\nu_0$$





Detailed balance revisited

“That leaves radiative recombination as the major [energy loss] process. Can this be avoided? The

answer is ~~no~~. ^{Yes!} If a radiative upward transition to generate the excitation is allowed, its reversal, the radiative downward transition ~~must be allowed as well.~~”

can be mitigated by breaking detailed balance via quantum coherence!



Summary

- Quantum coherence induced by an external microwave field can increase the quantum efficiency (open circuit voltage) of the photocell.
- Furthermore such a coherent driven photocell generates more **power**. The extra power produced by the device is much larger than that derived from the microwave source which creates the coherence.
- Induced coherence results in more efficient utilization of the pump photons by increasing absorption and/or quenching unwanted emission.



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Noise induced quantum coherence

- External source:
 - ✓ Lasing Without Inversion
 - ✓ Supercharged Quantum Heat Engine
- No external field
 - ✓ Fano interference (quantum noise)
 - ✓ Agarwal (Fano-Harris) lasing without inversion



“...The preceding coherent drive model illustrates the role of quantum coherence in a simple way. However, it is possible to generate coherence without the use of an external field. For example, quantum noise induced coherence via Fano coupling...”

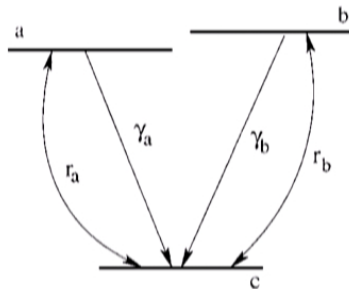


Noise induced coherence

PHYSICAL REVIEW A 74, 063829 (2006)

Inducing quantum coherence via decays and incoherent pumping with application to population trapping, lasing without inversion, and quenching of spontaneous emission

Victor V. Kozlov,^{1,2} Yuri Rostovtsev,¹ and Marlan O. Scully^{1,3,4}



Steady state coherence

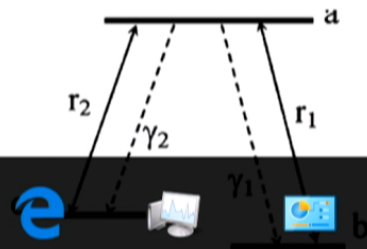
$$\rho_{ab} = \frac{p}{2} \frac{\sqrt{r_a r_b} - \sqrt{\gamma_a \gamma_b}}{r_a + r_b + \gamma_a + \gamma_b}$$

$$p \equiv \frac{\mu_{ac} \mu_{bc}}{|\mu_{ac}| |\mu_{bc}|}$$

Optics Communications 281 (2008) 4940–4945

Coherence induced by incoherent pumping field and decay process in three-level Λ type atomic system

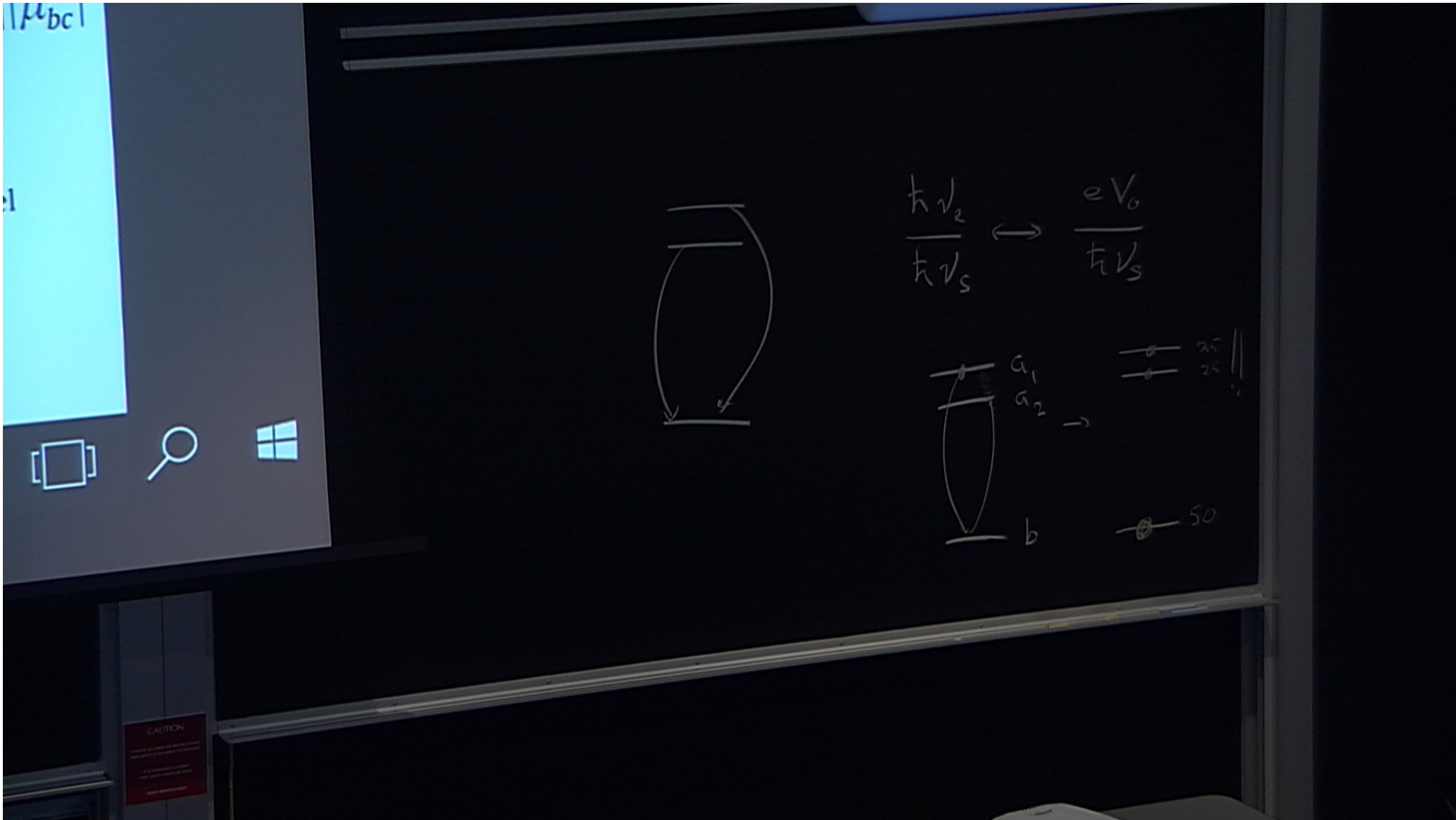
Bao-Quan Ou *, Lin-Mei Liang, Cheng-Zu Li



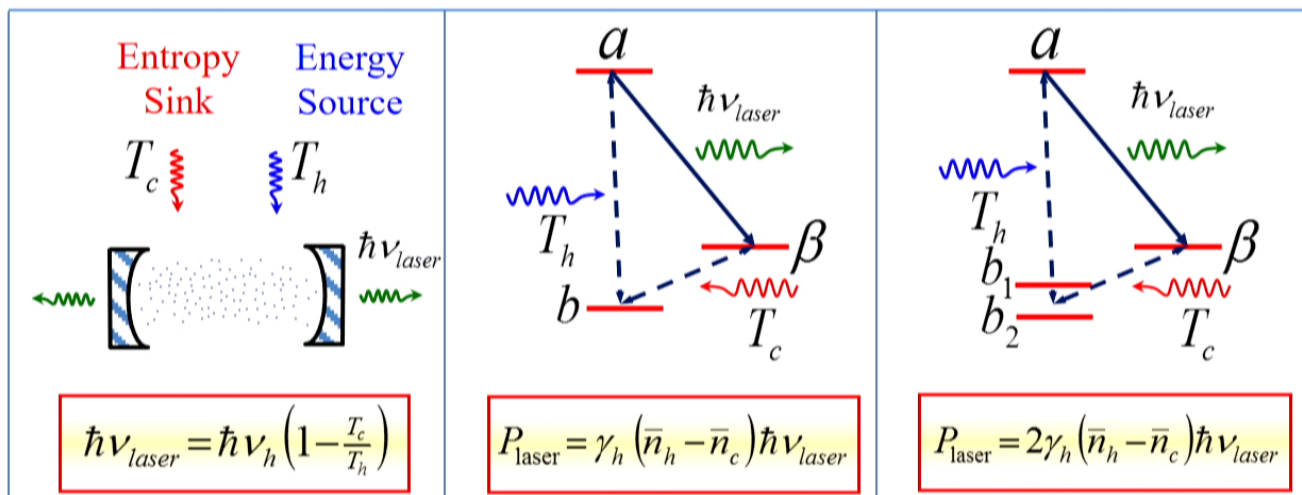
Steady state coherence

$$\rho_{bc} = - \frac{\sqrt{r_1 r_2}}{r_1 + r_2}$$

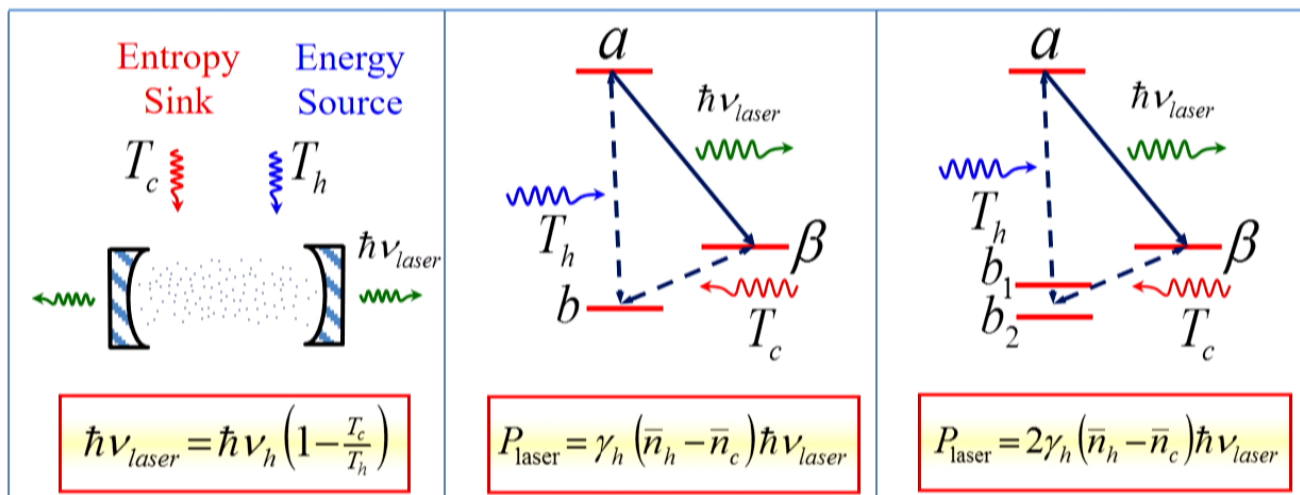




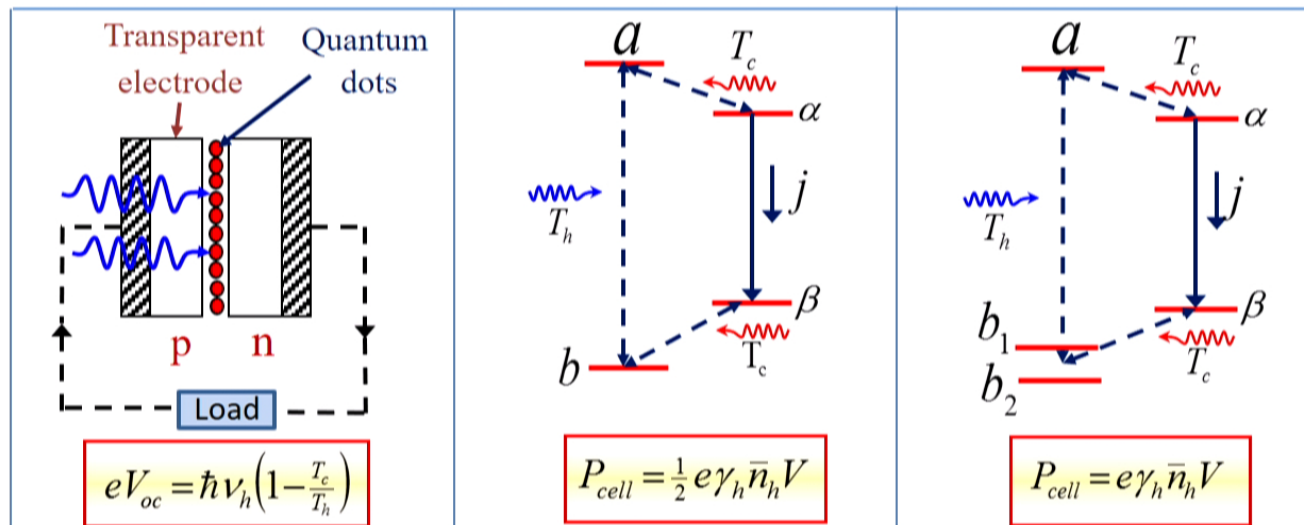
Noise induced quantum coherence can double the laser power at no extra cost!



Noise induced quantum coherence can double the laser power at no extra cost!



Noise induced coherence can double photocell power at no extra cost!



Noise induced coherence in photosynthetic systems

Konstantin Dorfman, Dmitri Voronine, Shaul Mukamel, and Marlan Scully



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