Quantum Dots by QED

Abstract—High quantum dot (QD) efficiency is currently explained by impact ionization that allows more than one exciton to be successively produced in the absorption of a photon having Planck energy greater than the band gap of the semiconductor. But impact ionization is not likely the cause of high QD efficiency because successive absorption takes time while experiments show the excitons are formed instantaneously. Alternatively, high efficiency in QDs may be explained by the quantum electrodynamics (QED) confinement of electromagnetic (EM) radiation during the absorption of the photon. Since QED suppresses EM radiations in a confinement having frequencies lower than its EM resonance, and since QDs are resonant at least in the vacuum ultraviolet (VUV), the infrared (IR) radiation emitted from the atoms in the QD is suppressed. But IR radiation depends on the atom temperatures, and therefore QED effectively forbids any increase in the temperature of the QD. Absent an increase in temperature, the EM energy of the absorbed photon may only be conserved by increasing its frequency and consistent with the QED constraint that forbids EM radiation in the QD having frequencies lower than its EM resonance, the frequency of the absorbed photon is increased to at least VUV levels - the process called cavity QED induced EM radiation. Spontaneously, the VUV radiation produces additional excitons consistent with the observed instantaneous increase in QD efficiency. Extensions are made to IR multiphoton excitation (MPE) of porous silicon (PS), multiphoton dissociation (MPD) of molecules, classical radiation heat transfer of IR gases, and production of visible (VIS) light at ambient temperature.

I. INTRODUCTION

The efficiency of nanocrystal (NC) semiconductors is the electrical power produced as a percentage of the absorbed radiation. Efficiency in the conversion of photons to excitons in bulk semiconductors is usually based on the one photon – one exciton rule for photons having Planck energy greater than the band gap, the upper limit determined [1] to be about 31 %. The highest QD conversion efficiency [2] sometimes called carrier multiplication (CM) is typically about 66 %. Currently, high QD efficiency is explained by impact ionization [3] that allows more than one exciton to be successively produced in the absorption of a photon with Planck energy greater than the band gap of the semiconductor. But impact ionization by successive absorption takes time while experiments [4] show the excitons are formed instantaneously. Further, impact ionization fails to explain [5] almost the same QD efficiencies found for the widely different CdSe and PbSe semiconductors.

Subsequently, the "coherent–CM" mechanism was proposed [6,7] as an alternative to impact ionization, but required very large Coulomb coupling that could not be experimentally [5] verified. This prompted the similar high QD efficiencies found for the quite different CdSe and PbSe semiconductors to be explained by the instantaneous generation of high rates of multiexcitons by *virtual* single exciton states, thereby obviating the need for either resonance between single-exciton and multiexcitons states or very large Coulomb coupling.

In this paper, high QD efficiency is explained by the QED confinement of EM radiation. Exotic *virtual* exciton states are not invoked. IR radiation produced by temperature increase within the QD from the absorption of laser photons is suppressed because the typical EM resonant frequency of the QD is beyond the VUV. Since IR radiation depends on atom temperatures, QED effectively forbids the QD from increasing in temperature. This means QDs remain at ambient temperature during and after QD luminescence. Absent any increase in temperature, the photon energy absorbed in the QD may only be conserved by an increase in the photon frequency. But QED also forbids EM radiation within the QD having frequencies lower than its EM resonance, and therefore the frequency of the photon is spontaneously increased to at least the VUV - the process called cavity QED induced EM radiation. By this theory, high QD efficiency is explained by the excitons produced under QED induced EM radiation.

Usually, QED induced EM radiation explains how photons or electrons are observed at ambient temperature, specifically in evacuated QED cavities momentarily or permanently embedded in a liquid or solid medium, e.g., flow electrification [8], and the Casimir effect [9]. However, cavity QED induced EM radiation is applied here to QDs in the solid state, the confinement of EM radiation in the QD only differing from that in evacuated cavities by its refractive index n_r , i.e., the dimension d of the fundamental optical mode standing across the QD is $d = 1 / 2 n_r$, where $n_r \sim 3.5$ for semiconductors. In contrast, the confinement dimension d of the fundamental mode in evacuated QED cavities is, d = 1 / 2, where $n_r = 1$. What this means is that in semiconductor QDs, the suppression of IR radiation occurs in a smaller confinement dimension d than in an evacuated QED cavity, but otherwise the process of QED induced EM radiation in the frequency up-conversion of suppressed IR radiation to the EM resonance of the QED confinement remains the same.

II. THEORY

The QED induced EM radiation process in a QD is illustrated in Figure 1. The QDs are taken to be spherical NCs of radius *R* having an EM resonance in the VUV. A laser photon having Planck energy E_P in excess of the band-gap E_{B-G} is absorbed in the QD. Planck energy E_{B-G} produces 1 exciton while the excess $E_P - E_{B-G}$ is converted to IR heating.

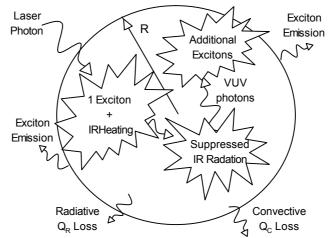


Figure 1 Laser induced Exciton Emission in QD

Subsequently, the IR heating tends to increase the QD atom temperatures and emit IR radiation. But QED suppresses EM radiation at IR levels in the VUV resonant QD, or equivalently, QED forbids the QD to increase in temperature. Absent increases in temperature, the EM energy of the absorbed photon is conserved by an increase in its frequency to the VUV resonant frequency of the QD. If the VUV radiation has Planck energy in excess of the band-gap, additional excitons are formed in the QD, thereby exceeding the one photon – one exciton rule for semiconductors in the bulk. But the number of additional excitons allowed in the QD is limited by the Pauli Exclusion Principle and electrostatic repulsion, the excess of which recombine to cause the VIS emissions.

A. Excitons Available in QD at Ambient Temperature

Before considering the formation of excitons in the QD illuminated with laser photons, it is instructive to consider the how the finite amount of thermal kT energy in the QD at ambient temperature T_{amb} alone may produce a momentary burst of excitons. The total thermal kT energy U available in the QD,

$$U = \frac{3}{2} N k T_{amb} \tag{1}$$

where, N is the number of atoms in the QD. The IR radiation emitted from an atom at ambient temperature T_{amb} follows the Einstein-Hopf relation [10] for the harmonic oscillator, of which the dispersion of average Planck energy E_{avg} with wavelength I at 300 K is shown in Figure 2.

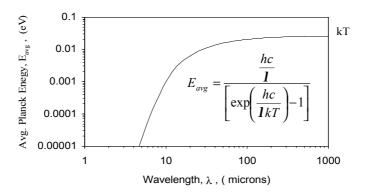


Figure 2 Planck energy of harmonic oscillator at temperature $T \sim 300$ K. In the inset, *h* and *k* are Planck's and Boltzmann's constants, and *c* is the speed of light.

Suppressed IR radiation in a QD may be understood by first considering an atom at ambient temperature T_{amb} placed in an evacuated spherical QED cavity of radius R having resonant wavelength I, where I = 4R. Figure 2 shows the IR radiation emitted from the atom absent confinement in free space, the full kT energy ~ 0.0258 eV is available for I > 100 microns; whereas, the kT energy available at I < 10 microns is insignificant. Thus, if the atom is placed in an evacuated QED cavity having R > 25 microns, its IR radiation is not suppressed, but if placed in an evacuated cavity having R < 2.5 microns, almost all its kT energy is suppressed. Similarly, semiconductor QDs of R > 7 microns and $n_r \sim 3.5$ suppress insignificant IR radiation. However, QDs of diameters d from 2 to 100 nm having EM resonant wavelengths from $I \sim 14$ to 700 nm suppress all thermal kT energy available in the atom at ambient temperature. Hence, IR suppression in QDs occurs at smaller confinement dimension d than in evacuated QED cavities.

The total IR energy suppressed in typical QDs at ambient temperature T_{amb} is,

$$U = 2\mathbf{p}kT_{amb} \left(\frac{R}{\mathbf{D}}\right)^3 \tag{2}$$

where, the atoms have 3 degrees of freedom, and **D** is the cubical spacing of atoms in the NC, $D \sim 0.3$ nm. Since the fundamental EM resonance is the lowest admissible frequency allowed within the QD, the suppressed

IR radiation is frequency up-converted to EM radiation having the Planck energy E_{VUV} ,

$$E_{VUV} = \frac{hc}{l} = \frac{hc}{4n_r R} \tag{3}$$

Conservation of EM energy gives the number N_{VUV} of VUV photons having Planck energy E_{VUV} ,

$$U = N_{VUV} E_{VUV} \tag{4}$$

$$N_{VUV} = \frac{U}{E_{VUV}} = 2p \left(\frac{R}{\Delta}\right)^3 \left(\frac{kT_{amb}}{E_{VUV}}\right) = 8pn_r R \left(\frac{R}{\Delta}\right)^3 \left(\frac{kT_{amb}}{hc}\right)$$
(5)

and,

The Planck energy E_{VUV} and number of N_{VUV} of VUV photons available at ambient temperature in QDs for $n_r = 3.5$ having radii from 1 to 100 nm are shown in Figure 3. Evacuated QED cavities are represented by $n_r = 1$.

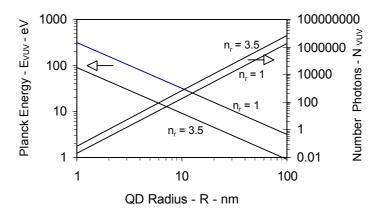


Figure 3 Planck energy and Number of Photons produced in QD

At the instant the QDs are formed, a single burst of VUV photons is produced from the thermal kT energy available at temperature T_{amb} . The QD thermal kT energy is then depleted and absent subsequent heating to recover ambient temperature, the QD resides in a state of very low thermal kT energy. However, the QD temperature is not lowered to absolute zero, but rather remains at ambient temperature. This can be understood from Fig. 2 that shows although the thermal kT energy of an atom vanishes in a VUV cavity, the temperature remains at ambient.

B. Steady Laser Induced Excitons

Lasers provide the direct means by which the QD atom temperature T_{amb} may be maintained while producing steady VUV radiation and attendant VIS light emission. In order to produce a steady rate dN_E/dt of excitons, the laser is required to provide photons at a rate dN_P/dt having Planck energy E_P in excess of the band-gap E_{B-G} . An upper bound to QD efficiency assumes there are no radiative Q_R and convective Q_C losses , the conversion of excess Planck energy $(E_P - E_{B-G})$ to excitons at steady state given by,

$$\frac{dN_P}{dt} \left(E_P - E_{B-G} \right) = \frac{dN_E}{dt} E_E \tag{6}$$

Providing $E_P > E_{B-G}$,

$$N_{E} = 1 + \frac{\left(E_{P} - E_{B-G}\right)}{E_{E}} \tag{7}$$

The fraction **h** of the full kT energy available in the QD at ambient temperature T_{amb} required to produce the exciton emission is,

$$\boldsymbol{h} = \frac{N_E E_E}{U} = \frac{N_P (E_P - E_E)}{2\boldsymbol{p} k T_{amb} (R / \boldsymbol{D})^3}$$
(8)

The total thermal kT energy U in the QD in terms of the radius R and the fraction **h** of the total U necessary to produce exciton emission having $E_E \sim 1.4$ eV for $N \sim 1$ and 6 is shown in Figure 4.

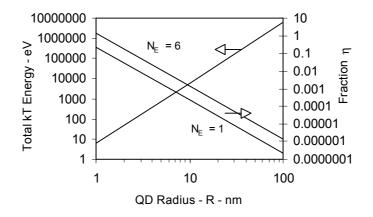


Figure 4 Total QD kT Energy and Fraction of Exciton Emission

The staircase efficiency of QDs (Fig. 13 of [7]) is reproduced here in Figure 5. Of interest is the QD efficiency threshold begins at values of (Photon energy $/ E_G$) > 2 and not 2.

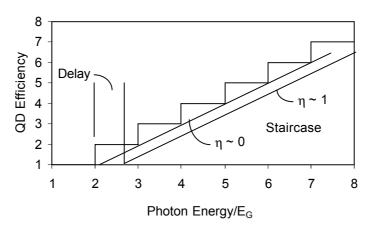


Figure 5 Delay in Staircase QD Efficiency

The delay in the threshold of QD efficiency h may be explained by the photon energy required to recover the thermal kT energy in the QD lost during the previous laser pulse. Fig. 4 shows the fraction h of the available kT energy in a QD necessary to produce N_E excitons decreases with increasing QD size. For small QDs having $h \sim 1$, the available thermal kT energy is low and after depletion, the laser provides most of exciton energy. Thus, (Photon energy / E_G) > 2 in Fig. 5. Indeed, the QD absent a laser resides in a state having minimal thermal kT energy, and therefore the laser must provide this energy before excitons are produced. Conversely, large QDs having $h \sim 0$ need little laser energy, and therefore Fig. 5 shows the QD efficiency staircase threshold begins at (Photon energy / E_G) ~ 2.

III. DISCUSSION

A. Thermal Radiative Emission or Photoluminescence from Porous Silicon?

Fundamental in the QED explanation of QD efficiency is the controversy [11] over whether the QD luminescence following laser illumination is thermal radiation (TR) or photoluminescence (PL). Here, TR is blackbody radiation depending on temperature, while PL is light emission in transitions between energy states.

QD luminescence begins in 1990 with the discovery [12] of luminescence from PS. The discussion of PS is relevant to QD luminescence because of the issue whether the PS luminescence is TR or PL. PS luminescence was first explained [12] by the quantum confinement of electrons in the thin pore walls, the surrounding evacuated pores acting as potential barriers. But this simple explanation for QED confinement is even now difficult to understand because of the obvious question:

How did the electrons become trapped in the PS pore walls?

Indeed, understanding how the electron becomes trapped in the PS walls is central to explanation of PS and QD luminescence. For QDs, the usual explanation [13] is that the laser creates free excitons, say in the GaAs surrounding the QD, the excitons then diffusing into the QD to relax down to the lowest energy level in the QD. But this is unlikely because the potential confining the electron within the QD would certainly block an electron external to the QD from entering and occupying the lower QD states. Similar arguments can be made against the electrons confined in PS. Clearly, another mechanism is operating to trap the electron in the PS and QDs, and it is plausible that the same mechanism would explain the formation of excitons in QDs. Here, cavity QED induced EM radiation is proposed as the mechanism by which the excitons are produced and electrons trapped in the in the QDs and PS.

Trapping the electron in PS or QDs proceeds spontaneously by QED induced EM radiation at the instant the temperature in a confined space tends to increase by heating whether directly from the laser or indirectly by convection from surrounding gases, or conduction from supporting material.

The electron does not migrate into the QDs, but is trapped after being created in the QD by VUV radiation by QED induced EM radiation.

Recently, a key experiment [11] was performed to determine whether QD luminescence was TR or PL. The experiment showed the QD luminescence in a vacuum was quenched by gas pressure. On this basis, it was concluded that QD luminescence cannot be PL because the transitions between energy states would have to depend on the pressure of the surrounding gas. Since quantum levels do not depend on gas pressure, QD luminescence was concluded not to be PL, and by default could only be TR.

But this argument is based on the premise that PL and TR are the only mechanisms by which QD luminescence may occur. Moreover, TR alone cannot be the direct cause of QD luminescence. Blackbody emission requires very high temperatures to sustain IR, VIS, and UV emissions, e.g., a temperature of 14,000 K is required to sustain a near IR photon at 1.4 eV. Since the QDs have to vaporize to produce the near IR photon, it follows that TR is also not the source of QD luminescence. However, TR increases the temperature of the bulk to enhance conductive heat transfer into the QD, the tendency for the QD to increase in temperature producing VUV radiation that: (1) by PL produces QD luminescence, and (2) by the photoelectric effect creates electrons in the QD.

The bulk QD or PS substrate may increase to high temperatures, but this not the cause of luminescence. Rather, it is the tendency of local confined QED regions to increase in temperature that produces the EM radiation which excites the vibration and electronic states. Bulk temperatures in QDs and PS are far lower than the blackbody temperatures necessary to directly excite the spectral peaks.

In 1992, PS luminescence was proposed [14] caused by quantum confinement in the MPE up-conversion of IR laser radiation in a double-resonance-enhanced nonlinear-optical process. The similar PL spectra found for PS irradiated with IR and UV photons was taken as confirmation of the high efficiency of MPE of IR photons in producing the equivalent UV photon in contradiction to questionable efficiency of MPE. Quantum confined IR radiation is also the basis for cavity QED induced EM radiation, but differs from [14] in that the IR radiation from the atom is confined and not that from an external IR laser.

QED induced EM radiation is a quantum confinement induced effect. But the quantum confinement is that of the IR radiation emitted by the atoms within the confinement, not by MPE of IR photons from a IR laser. By the QED theory, the IR laser is only of importance in PS or QD luminescence as the source of heat that tends to increase the temperature of the atoms within the QED confinement.

MPE is an inefficient mechanism for up-converting IR photons to the VUV; whereas, QED induced conversion is highly efficient.

Later in 1995, PS luminescence was proposed explained [15] by MPE. PL spectra excited by IR laser pulses at wavelengths from 0.532 to 4.9 microns were found similar to that obtained by UV excitation having Planck energy above the Si band gap, and therefore the MPE was taken to be confirmed independent of the photon energy. However, the MPE required 7 photons at 4.9 microns to produce a single 1.77 eV photon which is again questionable because MPE is known as an inefficient IR frequency up-conversion process.

QED induced EM radiation relies on the QED confinement to produce the VUV necessary for luminescence and does not depend on the IR photon energy. The IR photons simply tend to increase the temperature of the PS atoms in a locally QED confined regions, and thereby produce the luminescence.

Also in 1995, PS luminescence [16] from MPE using a CO_2 laser differed from [15] in that the PS was oxidized to form a very large number of SiO bonds in the surface. As in other MPE excitations of PS, PL intensity increased supralinearly with laser intensity. TR was dismissed as the cause of PS luminescence because the temperature and emission spectra did not depend on pulse energy, and instead the source was attributed to SiO surface states. In fact, vibrational excitation by MPE of the SiO molecule has been taken [11] as experimental support for PS luminescence by surface states. However, the PS luminescence including excited surface states may be readily be explained by the tendency of temperatures in QED confined spaces to increase.

MPE is related to vibrational states with well defined transitional energies, but PS luminescence is observed over a wide range of IR laser photon energies. This means the IR radiation simply increases the bulk PS temperature with local QED confined regions producing VUV radiation that by PL excites the PS including surface SiO states.

In 2000, PS luminescence in free-standing films [16] was thought caused by TR at temperatures measured at about 1000 K. In fact, the high temperatures are consistent with the hypothesis earlier advanced [17] that PS luminescence is not caused by any kind of exotic excitation process, but simply by TR. However, TR cannot explain the more recent [18] finding of C_2 emission from nano-carbon under microwave radiation in a vacuum.

B. Laser Dissociation of Molecules

The applicability of cavity QED induced EM radiation to QDs and PS as an alternative to MPE suggests that other areas of physics where MPE is a well established phenomenon [11] might also be similarly explained. One such area is the MPD of polyatomic and macromolecules by IR lasers. After all, a molecule like a QD or PS having a small number of atoms should follow the same physics.

In 1971, MPE was performed [19] with 10.6 micron CO_2 IR laser radiation of absorbing gases, e.g., NH₃, CCl_2F_2 , and C_3H_6 . The laser induced MPD luminescence was instantaneous followed by delayed sparking. The instantaneous phase was thought [20] to be caused by collisionless uni-molecular MPD of single molecules up the vibrational ladder, while the sparking was caused from collision-induced fragments. The C_2 Swan bands centered at 516 and 563 nm observed [19] in the IR laser induced luminescence from C_3H_6 suggested that by MPE the IR radiation would reveal the Swan bands in all hydrocarbon gases.

TR finds difficulty in producing detailed C_2 band structures, but the spectral bands are a natural consequence of PL from the QED induced EM radiation produced in the molecule upon IR radiation. Swan bands centered at 516 and 563 nm having Planck energies of 2.4 and 2.2 eV cannot be explained by blackbody temperatures of about 24,000 degrees because any solid state materials would vaporize.

Nevertheless, the issue of whether IR induced MPD of molecules is TR or PL continued [21] with the argument that before a molecule dissociates, the thermal energy from the IR excitation may be regarded as randomized in all its vibrational modes. But slow thermal heating with IR photons does not occur because QED forbids the molecule to increase in temperature.

In cavity QED induced radiation, IR heating of a molecule is conserved not by an increase in temperature, but by conversion to Planck energy at the frequencies of all vibration states. It is not thermal energy that is randomized in the vibration modes as currently thought, but rather the EM energy of the absorbed IR photon.

In the rapid MPD of a molecule under IR radiation, the Planck energy that occurs instead of a temperature increase causes the molecule to dissociate as if irradiated by EM radiation in excess of its dissociation energy. However, this is not the MPD thought to occur by MPE of TR. For example, the IR laser MPD of silylene [22] showed detailed SiH₂ structure near 17,250 cm⁻¹ or 2.14 eV that cannot be explained by MPE, but rather by the spontaneous conversion of absorbed IR photons into all quantum states by QED induced EM radiation.

Conversely, an argument may be made that the laser induced luminescence [23] from gas phase C_{60} and C_{70} fullerenes is TR because the fullerene spectra has the same shape as that for a tungsten filament at 3000 K, although the tungsten produces a far brighter light. Absent C_2 emission, the fullerene spectra are essentially those of blackbody radiation at 3000 K. In contrast, the C_2 bands from the microwave irradiation of nanocarbon [18] are observed as the temperature reached 3915 K. But blackbody radiation cannot explain the C_2 bands. It is likely the nanocarbon substrate at 3915 K is emitting blackbody radiation and adjacent local QED confinement regions are producing the C_2 bands by cavity QED radiation.

C. Classical Radiative Heat Transfer

Classical radiative heat transfer [24] is the study of the interchange between IR absorbent gases, e.g., CO₂, CO, and H₂O and the walls of an enclosure. By treating the enclosure as a broadband blackbody laser from ~ 1 to 20 microns, frequency up-conversion is generally necessary to excite the molecules, in particular the near IR vibration states. Similar to IR laser excitation, the gases may be excited with the blackbody laser by MPE up the vibration ladder. But again, MPE is an unlikely mechanism for up-converting blackbody radiation from the enclosure walls because of inefficiency, and therefore QED induced radiation provides a credible explanation of how gases are excited in radiative heat transfer. Clearly, EM radiation and not the temperature of the enclosure walls may excite the vibration state of a molecule, although temperature defines the EM radiation available in the enclosure wall. For example, the blackbody radiation spectrum at 1100 K that envelopes (Fig. 23.22 of [24]) the peak of the emission spectrum of H₂O and CO₂ does not mean a temperature of 1100 K excited the vibration states. Rather, the H₂O and CO₂ molecules in the same enclosure at another temperatures, say 700 K, are also excited, although at a lower intensity than at 1100 K. In effect, QED induced radiation excites all vibration states of the IR absorbent molecules at the instant the blackbody radiation is absorbed with intensity that increases with enclosure temperature.

IR absorbent gases may be excited by cavity QED induced EM radiation by blackbody radiation from the walls of an enclosure. To increase the frequency of blackbody radiation from the enclosure walls to the frequency of the vibration states, QED induced radiation is required, as MPE is inefficient.

D. Light at Ambient Temperature

Lasers are not necessary to produce light from the QD. Indeed, VIS light may be produced at ambient temperature from the blackbody TR available in the natural surroundings. Recall that the burst of excitons produced at the instant a QD is formed leaves the atoms in a VUV confinement with very low thermal kT energy, but their temperature is still ambient. With both the QD and the ambient at the same T_{amb} temperature, heat flow is precluded by the 2nd law of thermodynamics. But the thermal kT energy of an atom is its kinetic energy, and therefore the velocity of the atoms in the QD is low compared to that of the air molecules in the ambient striking the QD surface. Without violating the 2nd law, the QD luminescence is caused by the transfer of kT energy from the air molecules to the atoms in the QD.

IV. SUMMARY AND CONCLUSIONS

- QDs and PS are excited by QED induced EM radiation if a steady source of heat is provided. Otherwise, the QD and PS only produce a burst of excitons as they are first formed at ambient temperature.
- Following the first burst of excitons, the thermal kT energy of the atoms within the QED confinement is reduced to insignificant levels, but the temperature remains near ambient. Suppressed IR does not lower the atoms in the QED confinement to absolute zero. Heat must be supplied to recover the thermal kT energy of the QED confinement.
- Absent lasers, the kT energy in QDs or PS may be sustained from ambient blackbody TR in the surroundings, although the VIS light emission is low level.
- The transfer of an external electron into PS or QDs does not occur by diffusion through the same barrier that eventually confines it in the PS or QD. Instead, the electron is trapped as a consequence of QED induced EM radiation at the instant heat tends to cause the PS or QDs to increase in temperature.
- Laser photon frequency is increased upon absorption to the EM resonant frequency of the QD or PS, thereby exciting the semiconductor to produce excitons.
- TR is not the source of QD or PS excitons, although TR provides the heat to continually recover the QD or PS temperatures in steady VIS light production.
- Blackbody radiation cannot explain the detailed structure of Swan bands found in the dissociation of hydrocarbon gases under IR radiation unless it is frequency up-converted.
- Similar to QDs and PS, molecular dissociation by IR lasers does not occur by MPE. Instead, laser induced MPD occurs by the collisionless process of cavity QED induced EM radiation.
- Cavity QED radiation supports the classical theory of radiative heat transfer of IR absorbent gases in an enclosure by increasing the frequency of blackbody EM radiation from the enclosure walls necessary to excite the higher level molecular vibration states.
- Low level light may be produced at ambient temperature without violating the 2^{nd} law in the collisional transfer of kinetic energy from molecules having high kT energy in the surroundings with the surface of the QDs or PS.

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