A complete introduction through up conversion photoluminescence and its mechanisms

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Abstract

In this context a very mature and precise review is done to the photoluminescence up-conversion phenomenon. After an introduction to the photoluminescence up-conversion, the mechanisms are studied. Furthermore, the factors which are effective in the enhancement of photoluminescence up-conversion are indicated and analyzed. The conclusions of our studies show that the removal of the linear momentum's conservation bond in the heterostructures has a very prodigious effect on the efficiency of this optic phenomenon.

Keywords: Anti-Stokes Photoluminescence, Electro-optical effects, Optoelectronics.

Resumen

En este contexto se hace una revisión muy madura y precisa del fenómeno de up-conversion de la fotoluminiscencia. Después de una introducción a la conversión ascendente de fotoluminiscencia, se estudian los mecanismos. Además, se indican y analizan los factores que son efectivos en la mejora de la conversión ascendente de fotoluminiscencia. Las conclusiones de nuestros estudios muestran que la eliminación del enlace de conservación del momento lineal en las heteroestructuras tiene un efecto muy prodigioso en la eficiencia de este fenómeno óptico.

Palabras clave: Fotoluminiscencia Anti-Stokes, Efectos electro-ópticos, Optoelectrónica.

I. INTRODUCTION

Sometimes due to the shortage of the instruments for exciting the studied sample, we don't have a laser with the frequency more than the band gap of the sample. Then we must use a lower-frequency laser. The manner of performing this process leads to the up-conversion photoluminescence (PL) and studying the effective mechanisms of it. Up-Converted PL is observed in the various systems such as atoms and molecules [1], polymers [2], amorphous semiconductors [3], and the crystal semiconductors with the heterostructures. The generation of emission energy with an increasing of about 700 (mev) compared to the excitation energy of GaAs/ordered-(Al) GaInP system [4] which includes the red, orange and green areas of the spectrum is one of the reasons of studying this phenomenon, which is so important in terms of technology. Diagnosis of the type of band alignment in the interface of heterostructures is indicated as another reason. Furthermore, this phenomenon is utilizable in frequency conversion of the semiconductor lasers.

II. MATERIALS AND METHODS

Photoluminescence Up-Conversion Phenomenon

In the up-conversion PL or anti-Stokes photoluminescence (ASPL) phenomenon, the energy of the exciting descending photons is lower than the dispatched photons, the process of obtaining lower energy by the sample and dispatching more energy is known as energy up conversion. If the energy of the descending photon would be surmised as hν , then the electron of the valence band will extend itself into a visual level in the band gap and then it will extend itself onto the edges of conduction band by the proper mechanism which is explained. At the next stage, the light emitting recombination will be occurred between the electron and the valence band located hole. In general, the difference between normal photoluminescence (NPL) and ASPL is that the NPL has the lower excitation energy. Furthermore, a pure heat is generated in the sample semiconductor, because the energy of the descending photons is more than the band gap. Certainly, a part of that energy is consumed for the vibration of the network. But, the ASPL is followed by the absorption of heat in the semiconductor.

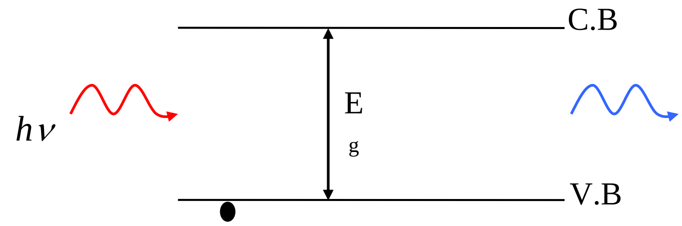

FIGURE 1. A representation of the ASPL phenomenon; energy of thedescending photon is lower than the band gap by an amount of $h\nu$ ($h\nu < E_g$).

The effective factors in increasing the efficiency of the ASPL are listed below.

A) The permission of removal of the linear momentum's conservation bond of carriers by the presence of a heterogeneous interface: It is obvious that the existence of heterogeneous interface has fundamental effects on the behavior of the energies and carrier wave functions in the quantum systems. The permission of the linear momentum of electrons in the vertical horizon of heterogeneous interface is distinct by theoretical calculations [6].

In fact, the electrons excited by the laser source are ejected in the horizon which the sample has the most region heterogeneity. Hence, the efficiency of light emitting recombination is increased by removing the linear momentum's conservation bond.

B) The existence of substitutive or boundary conditions in the semiconductor with the lower band gap:

In the heterostructures after excitation of carriers and when they reached into the edges of conduction band, the existence of substitutive conditions for preventing the carriers to return and light emitting recombination in the lower band gap semiconductor area is of important essence, so the carriers reach themselves to the longer band gap semiconductor by absorbing the next photon.

C) The existence of the type-II band alignment in the heterostructure:

As illustrated below, the type-I heterostructure, both electron and hole must overcome the dam energy and reach themselves to the longer band gap semiconductor area, however, in the type-II heterostructures, energy conversion of one kind of carriers is efficient.

In fact, for observing the ASPL signal, absorption of three photon in the type-I heterostructure, and two photons in the type-II are essential.

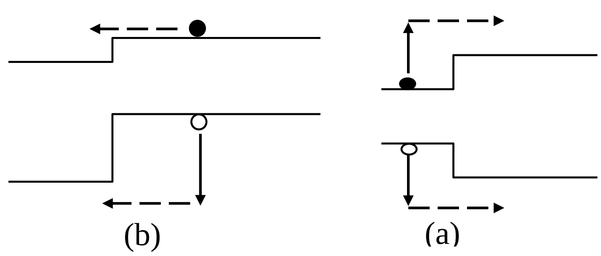

FIGURE 2. A representation of energy up-conversion for a) type-I heterostructure b) type-II heterostructure.

D) The existence of band fluctuations (substitutive conditions in the conduction band) prevents from the returning of carriers to the lower band gap, in the larger band gap semiconductor area, in the first type of heterostructures.

A. Photoluminescence Up-Conversion Mechanisms

The mechanisms which are often considered for the justification of energy up-conversion are mentioned below.

1) One step- two photon absorption (TPA)
2) Two step -two photon absorption (TS-TPA)
3) The Auger Process

B. One Step Two Photon Absorption (TPA) Process

The TPA process is explained by absorbing two photons in one step, totally coherently, and on the assumption that there is a middle state in the sample. The descending photons are overlapped in the space and especially in the time. Furthermore, each photon's energy is lower than the band gap's energy. Thus, the middle state in the sample is not real and the lifetime length of an excitation in this virtual position is defined by the time interval between energy up-conversion and the next real position.

In the first step, the electron in the valence band reaches itself onto the virtual band by absorbing the descending photon, Afterwards; the electron is transferred to the conduction band. The light emitting recombination between electron and the hole in the valence band results in the photon which has energy size of band gap, equal to sum of two absorbed photons. The TPA is considered as a nonlinear process due to the coherent absorption of two photons.

In general, the equation of light intensity variation in the semiconductor for absorbing two photons is

$$\frac{dI}{dZ} = -\beta I^2 \cdot$$

In this equation β is the coefficient of absorption of two photons which is surmised to be constant, and Z is the direction of laser emission. The above equation expresses that laser intensity variation has quadratic dependence on laser intensity, which is expected for two photons.

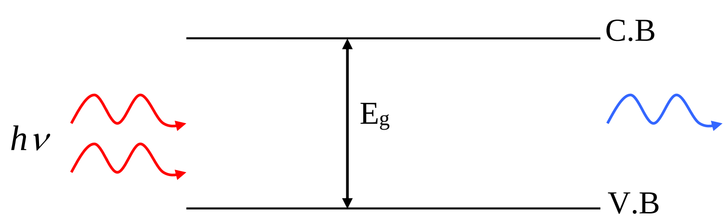

FIGURE 3. A representation of TPA process.

C. Two Step Two Photon Absorption

In TS-TPA process, absorption of two photons is occurred in two steps. The major differences between TS-TPA and TPA is: The middle state in this process, unlike the TPS process, is real. Furthermore, in the second step, the recombination of the excited carriers may be occurred before the absorption of the second photon.

In the TS-TPA process in the type-II heterostructure, in the first step electron absorbs a photon and reaches itself near the interface of heterostructure, and an exciton is generated in the lower band gap semiconductor. Afterwards, the next photon is absorbed by the hole in the valence band of lower band gap semiconductor and the hole obtains the sufficient energy to diffuse into the valence band of the bigger band gap semiconductor. Eventually, a light emitting recombination is occurred between the hole and the electron in the conduction of the bigger band gap semiconductor.

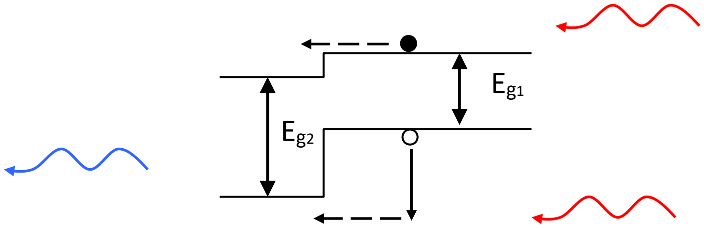

FIGURE.4. A representation of TS-TPA in the type-II heterostructures.

But, this process is occurred in the type-I heterostructure by absorbing three photons in three stages. It is noticeable that energy of descending photons is more than the lower band gap semiconductor, but less than the bigger band gap semiconductor

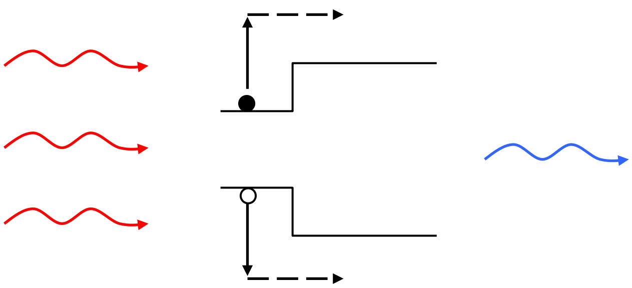

FIGURE 5. A representation of TS-TPA process in the type-I heterostructure.

In the TS-TPA process, the second absorption stage can be occurred by absorbing the sample's photon. This photon is called recycling photon, which is generated by the recombination between electron and hole in the semiconductor area, and is not related to the exciting laser. This photon could be absorbed by the laser in the second stage, thus the energy increasing equal to the photon's energy must be observed in the ASPL signal.

D. Auger Process

In the Auger process, the energy and momentum of a non-radiative recombination of the electron and hole couple is transferred to the next electron or hole, afterwards, this energy and momentum is converted to the dispatched photons. As illustrated below, in the first picture, electron is excited, but in the second picture, the hole is created in the cleavage of the valence band by Auger process.

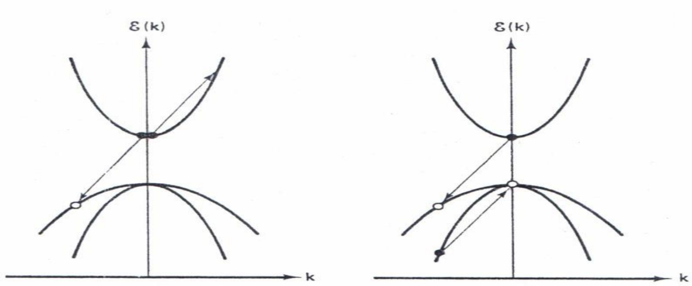

FIGURE 6. A representation of Auger process. a) Energy and momentum of recombination is given to an electron. b) Energy and momentum of recombination is given to a hole.

Auger process is known as Auger in heterostructures, because linear momentum conservation is removed and there is no heat threshold in this process. In this model, laser photons create electrons and holes in the lower band gap semiconductor area. These carriers are sent into the edges of the conduction band of the lower band gap semiconductors, then, one electron recombines with one hole and the produced energy is transferred into another electron or hole by recombination (Auger process), thus, electrons or holes are inverted and sent into the edges of bigger band gap semiconductor. Afterwards, they are trapped in the semiconductor area owing to the presence of localized mechanisms and after recombination in this area, the anti-Stokes PL is generated.

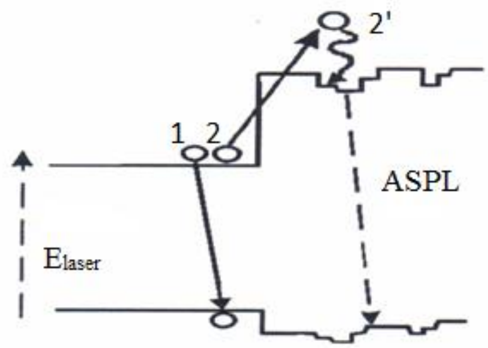

FIGURE 7. A representation of the Auger process.

Auger process is of a low efficiency in the bulk structures due to the imposition of linear momentum conservation bond, but in the heterostructures the linear momentum conservation bond is removed in the vertical direction. Thus, the transmission of the inverted carriers from lower band gap into the bigger band gap becomes easier. Hence, the Auger process in the heterostructures has a significant efficiency rather than in bulk structures.

E. The relation between anti-Stokes PL intensity and excitation power

Principally, the relationship between anti-Stokes PL intensity and excitation power is measured for more study in the major mechanisms of energy up-conversion. Anti-Stokes PL intensity is related to the excitation power by the equation of

$$I_{ASPL} = CP_{exc}^{\beta},$$

where P_{exc} is the excitation power.

The TPA process mostly happens in the $1000\,{}^{w}\!/_{cm^2}$ or more order of excitation power. But in TS-TPA process, I_{ASPL} is proportional to the number of second absorption stage photons and N_{exc} (boundary exciton states related to the impurity and deep localized states).

The number of photons absorbed in the second stage (N_{ph}) is directly commensurate with the excitation laser intensity. In low excitation intensities, the middle condensed states (N_{exc}) are still not saturated; I_{ASPL} is directly proportional to excitation laser intensity.

Since I_{ASPL} is proportional to $N_{ph}N_{exc}$, excitation laser intensity has quadratic dependence on I_{ASPL}. But in bigger intensities, the middle states are saturated, which means (N_{exc}=N_{ph}), the I_{ASPL} is proportional to N_{exc}=N_{ph} and consequently to $I_{ASPL.}$ Thus, anti-Stokes PL intensity probably is increased quadratic in lower excitation power and linear in bigger excitation power. In Auger process, I_{ASPL} is proportional to excitation power with third power ($\beta \cong 3$), because three electrons and three holes are required to create an excited couple of electron-hole.

Furthermore, I_{ASPL} has quadratic dependence on excitation power, when excitation efficiency of electrons is bigger than holes. It is also essential to say that Auger process is not accepted in low excitation powers, because the carrier wave functions overflow is essential for Auger recombination.

VI. CONCLUSIONS

In this article, we have introduced Anti-stokes photoluminescence process, which is very important at semiconductor's industry, and we have studied mechanisms, which are used for explanation this process. In our opinion, this article can be useful for both theoretical and experimental research in this field.

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