Thermoluminescence from Silicon quantum dots: A model study

Nebiyu Gemechu Debelo[1] and S. K. Ghoshal[2]
[1]*Department of Physics, Mizan-Tepi University, Faculty of Natural and Computational Science, Tepi, Ethiopia.*
[2]*Addis Ababa University, Department of Physics, Addis Ababa, Ethiopia.*

E-mail: nafsif@gmail.com

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Abstract

Most of the bulk materials start showing fascinating behaviors as their size is reduced to nano level. The ability to emit light as a consequence of absorption of energy (luminescence) is one of these exotic behaviors. There are different types of luminescence observed from nano silicon. Thermoluminescence (TL) is the emission of light from an insulator or semiconductor when it is heated. It is the thermally stimulated emission of light due to absorption of energy from radiation. So, there is a direct relationship between temperature (due to absorption of heat or radiation) and the TL intensity. We develop a model for calculating the TL intensity as a function of temperature and dot size. Our results are quite new and require future studies.

Keywords: Thermoluminescence, recombination, trapping, detrapping, quantum confinement.

Resumen

La mayoría de los materiales empiezan mostrando comportamientos fascinantes como su tamaño se reduce a nano escala. La capacidad para emitir luz como consecuencia de la absorción de energía (luminiscencia) es uno de estos comportamientos exóticos. Hay diferentes tipos de luminiscencia observada desde nano silicón. Termoluminiscencia (TL) es la emisión de luz de un aislante o semiconductor cuando es calentado. Es la emisión de luz estimulada térmicamente debido a la absorción de energía de la radiación. Por lo tanto, existe una relación directa entre la temperatura (debido a la absorción de calor o radiación) y la intensidad de TL. Desarrollamos un modelo para calcular la intensidad TL en función de la temperatura y el tamaño de punto. Nuestros resultados son bastante nuevos y requieren estudios futuros.

Palabras clave: Termoluminiscencia, recombinación, captura, recaptura, confinamiento cuántico.

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I. INTRODUCTION

Thermoluminescence is the emission of light from an in insulator or semiconductor when it is heated, that is it is the thermally stimulated emission of light following the previous absorption of energy from radiation [1, 2]. There are three essential ingredients necessary for the production of thermoluminescence [1]. Firstly, the material must be an insulator or semiconductor-metals do not exhibit luminescent properties. Secondly, the material must have at some time absorbed energy during exposure to radiation. Thirdly, the luminescence emission is triggered by heating the material. In addition, there is one important property of thermoluminescence. It is a particular characteristic of thermoluminescence that, once heated to excite the light emission, the material cannot be made to emit thermoluminescence again by simply cooling the specimen and reheating. In order to re-exhibit the luminescence, the material has to be re-exposed to radiation, whereupon raising the temperature will once again produce light emission [1]. The fundamental principles which govern the production of thermoluminescence are essentially the same as those which govern all luminescence processes, and in this way the thermoluminescence is one of a large family of luminescence phenomena. Luminescence can result because of two cases. One is due to the recombination of free electrons with trapped holes and the other is due to the recombination of free holes with trapped electrons. But in our model we have worked for the first case *i.e.* luminescence due to the recombination of free electrons with trapped holes.

II. MODEL CALCULATION

Under this model, we first calculate the number n of detrapped (free) electrons per unit volume to obtain the thermoluminescence intensity as function of temperature.

At equilibrium,

$$n = V\int D_e(\epsilon) f_0(\epsilon, T) d\epsilon. \quad (1)$$

where V is the volume [3].

$$f_0(\epsilon, T) = \frac{1}{exp\frac{\epsilon-\epsilon_f}{K_B T}+1}, \quad (2)$$

and

$$D_e(\epsilon) = \frac{1}{2\pi^2}\left(\frac{2m_e}{\hbar^2}\right)^{\frac{3}{2}}\sqrt{\epsilon - E}. \quad (3)$$

where K is the wave vector, K_B is the Boltzmann constant, T is the absolute temperature and ϵ_f is the fermi energy. For detrapped electrons,

$$\varepsilon = E + \frac{\hbar^2 K^2}{2m_e}, \quad (4)$$

where m_e and E are the usual effective mass of electron and trap depth respectively.

Since the function $f_0(\epsilon, T)$ sharply depends on ϵ grows, it is possible to substitute infinity for the upper limit of integration.

$$n(T) = \frac{1}{2\pi^2}\left(\frac{2m_e}{\hbar^2}\right)^{\frac{3}{2}}\int_E^\infty \frac{\sqrt{\epsilon-E}}{exp\frac{\epsilon-\epsilon_f}{K_B T}+1} d\epsilon, \quad (5)$$

$$n(T) = V\frac{\sqrt{\pi}}{2}\frac{1}{2\pi^2}\left(\frac{2m_e}{\hbar^2}\right)^{\frac{3}{2}}(K_B T)^{\frac{3}{2}} \exp\left(\frac{-(E-\epsilon_f)}{K_B T}\right). \quad (6)$$

Since thermoluminescence is due to the annihilation of detrapped (free) electrons with trapped holes at trap center, the thermoluminescenc intensity I_{th} is directly proportional to the recombination rate. But the recombination rate is directly proportional to the number n of detrapped electrons; hence I_{th} is directly proportional to the number of free electrons n. Hence:

$$I_{th} \sim n. \quad (7)$$

Another important idea is that the detrapped electrons may follow two path ways (radiative and non-radiative path ways).But it is electrons that follow radiative path way that contribute to the thermoluminescence intensity; hence we can conclude that I_{th} is also directly proportional to the rate of radiative recombination R_{rad}.

$$I_{th} \sim R_{rad}\, n. \quad (8)$$

Once more I_{th} is directly proportional to the probability per unit time p of the release of an electron from the trap.

$$I_{th} \sim p R_{rad}\, n. \quad (9)$$

But $p = s\exp\left(-\frac{E}{K_B T}\right)$, where s is the trapping parameter, and for quantum dot (approximately spherical) of well defined size distribution, $A = 4\pi r^2$, where r is the radius of the dot. But it is found that s varies with temperature like T^{-b}, where -2 < b < 2 [1].

Moreover for quantum dot of well defined size distribution, $V = \frac{4\pi r^3}{3}$, where r is the radius of the dot. Using (5) and (9),

$$I_{th} \sim s\exp\left(\frac{-E}{K_B T}\right) R_{rad}\frac{4\pi r^3}{3}\frac{\sqrt{\pi}}{2}\frac{1}{2\pi^2}\left(\frac{2m_e}{\hbar^2}\right)^{\frac{3}{2}}(K_B T)^{\frac{3}{2}}\exp\left(\frac{-(E-\epsilon_f)}{K_B T}\right). \quad (10)$$

This equation can approximately be written as:

$$I_{th} \sim s R_{rad}\frac{4\pi r^3}{3}\frac{\sqrt{\pi}}{2}\frac{1}{2\pi^2}\left(\frac{2m_e}{\hbar^2}\right)^{\frac{3}{2}}(K_B T)^{\frac{3}{2}} exp\left(\frac{-2E}{K_B T}\right), \quad (11)$$

$$I_{th} = C_2 R_{rad}\, d^3 exp\left(\frac{-2E}{K_B T}\right). \quad (12)$$

where C_2 is constant and d is the diameter of the dot. This equation gives the thermoluminescence intensity as function of temperature and dot size.

III. RESULTS AND DISCUSSION

A. Results of TL intensity versus temperature

Under this model, we directly calculated the number n of detrapped (free) electrons per unit volume (at equilibrium) to obtain the thermoluminescence intensity as function of temperature. For a fixed value of b (which is approximately equal to -1.5) the thermoluminescence intensity has a peak value at a temperature of 50K and starts decreasing after this temperature.

In this model we have worked for 1-300K temperature range and found out that our results are in good agreement with the experimental results [4]. As it can be seen from the graphs ((a), (b) and (c)), the TL intensity increases with temperature and attains its maximum value near 50K and slowly decreases after this temperature. However, as compared to the experimental result (d), the TL intensity obtained from this model decays much slower after a temperature of 50K. This theoretical model is also in good agreement with the experimental results for temperatures lower than 1K. For such lower temperatures, the exponential term approaches zero and hence the TL intensity drops to zero. The fact that the TL intensity increases following the reduction in nanoparticle size shows that quantum confinement effect is playing major role in allowing direct band to band radiative recombination.

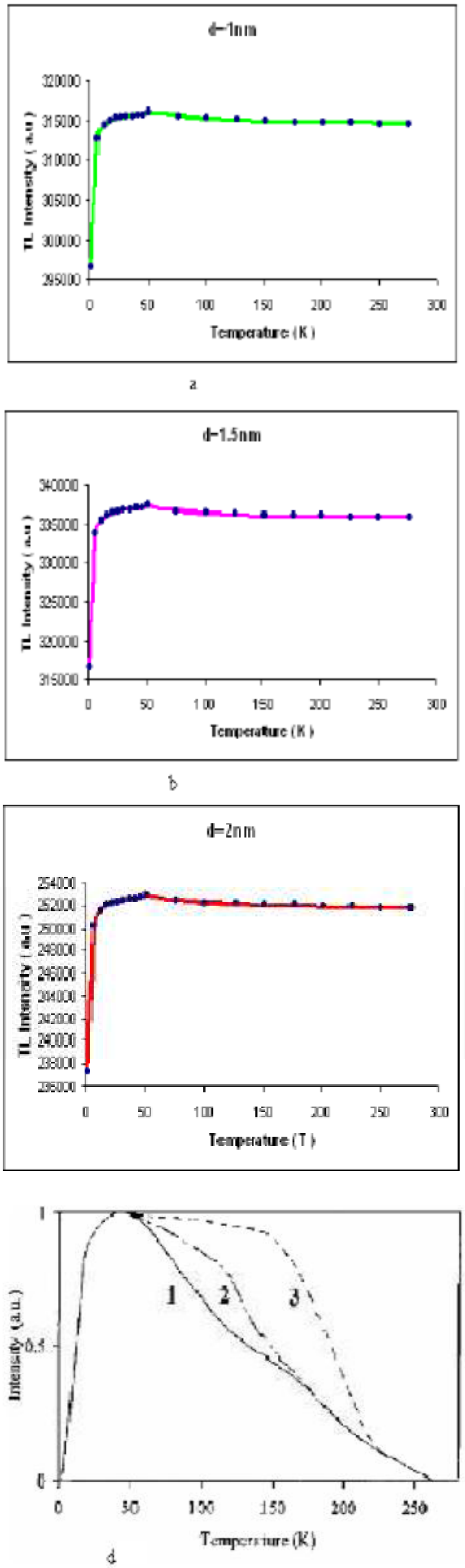

FIGURE 1. The calculated TL intensity for 1nm (a), 1.5nm (b), and 2nm (c) size samples. The experimental plot is also shown (d).

B. Results of TL intensity versus size

We are also interested in calculating the TL intensity as a function of the size of the dot for a given temperature. As expected, we observed that for a given temperature, the TL intensity increases with decreasing the size.

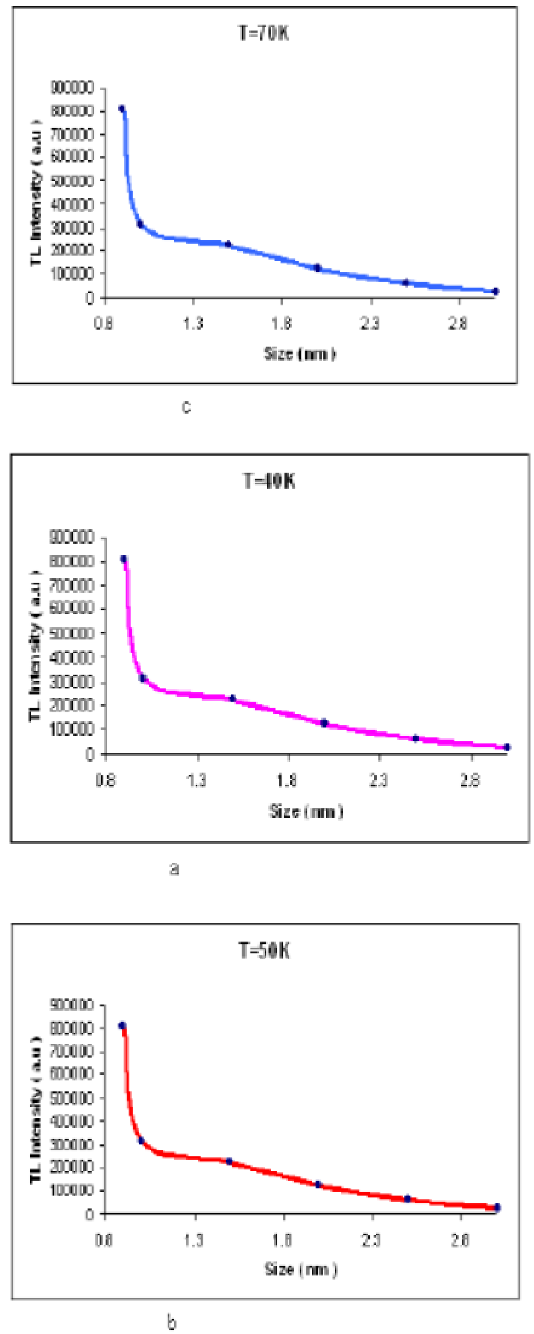

FIGURE 2. The calculated TL intensity versus size at different temperatures.

This is due to the quantum confinement effect which allows direct band to band radiative recombination following the widening of band gap. From this result one can easily understand that the TL intensity increases with increasing the band gap of the dot; because the reduction in size allows the increment of band gap.

IV. CONCLUSION

We have developed a powerful model for calculating the temperature dependent TL intensity. Of course the model has some limitation. We couldn't observe the fast decay mechanism of the TL intensity after 50K as observed from the experimental result.

We have also calculated the size dependent TL intensity and observed that it increases with decreasing the size of the dot. This is because of the fact that decreasing the size changes the band gap from indirect to direct and increases band to band radiative recombination. The results we obtained from the our model calculation can be explained well by the quantum confinement effect which allows band gap widening and direct band to band radiative recombination following the reduction in size. Our results are quite new and can also be applied to Ge, GaAs etc nanoparticles.

In general we have observed that the temperature dependence of the TL intensity is much similar to that of the PL and EL intensities. This shows that the mechanism of luminescence in the three types of luminescence is in general similar to one another.

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