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Conference Paper · December 2012

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Citation: AIP Conf. Proc. **1512**, 204 (2013); doi: 10.1063/1.4790982
View online: http://dx.doi.org/10.1063/1.4790982
View Table of Contents: http://proceedings.aip.org/dbt/dbt.jsp?KEY=APCPCS&Volume=1512&Issue=1
Published by the American Institute of Physics.

Observation Of Excitation Wavelength Dependent Photoluminescence From ZnO Nanoparticles Embedded In Mesoporous Silica

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Abstract. This manuscript reports on the excitation wavelength dependent photoluminescence (PL) of ZnO nanoparticles embedded in mesoporous silica (MS) matrix. FT-IR spectra of MS and ZnO/MS nanocomposite confirmed the unaltered structure of MS with ZnO loading. The successful encapsulation of ZnO nanoparticles in MS matrix was evidenced by HR-TEM. The PL of ZnO/MS was red shifted from 395 nm to 504 nm only by changing the excitation wavelength. This kind of unusual behavior is termed as red edge effect (REE).

Keywords: ZnO nanoclusters, mesoporous silica, photoluminescence, red edge effect.
PACS: 78.55.Et, 78.55.Mb, 78.67.Ef, 78.30.Fs.

INTRODUCTION

ZnO is one of the II-VI compound semiconductors which has been studied extensively for the past seven decades. The interest in the research on ZnO is due to its wide and direct band gap of 3.37 eV and large exciton binding energy [1]. The large exciton binding energy promises the efficient exciton emission at room temperature. Due to its unique properties, it finds applications in blue lasers, solar cells, varistors, sunscreens, liquid crystal displays, spintronics etc. Inspite of the decades of research, the origin of visible luminescence of ZnO nanoparticles is in dispute. In general, ZnO exhibits weak UV emission and strong and broad green emission ranging from (500-530 nm). Literature evidenced the numerous studies on PL of ZnO but studies of PL with variable excitation wavelength have been in scarce. Only few reports are available on excitation wavelength dependent PL of ZnO nanoparticles [1-4]. This kind of excitation wavelength dependent PL spectra attributed to different phenomenon such as quantum confinement of the amorphous ZnO nanoparticles [1], salvation and energy transfer in the case of nano-ZnO colloids [2] and amorphous structure and surface capping of ZnO nanoclusters [3].

In this work, very small ZnO nanoparticles are incorporated in MS successfully. With variation in excitation wavelength, PL spectrum red shifted continuously.

EXPERIMENTAL PROCEDURE

The preparation of MS and ZnO/MS nanocomposite is same as the procedure described elsewhere [5]. The prepared samples were annealed at 550°C at a rate of 2°C/min. The structure and morphology of the samples was studied by FT-IR and HR-TEM. PL measurements were carried out on Jobin Yuon spectrofluorometer.

RESULTS AND DISCUSSION

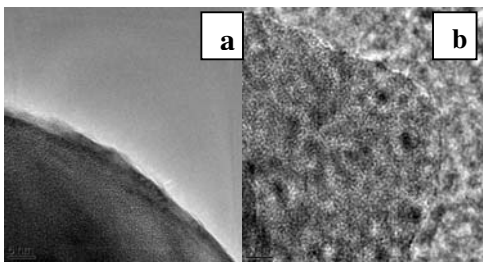

FIGURE 1. HR-TEM images of MS (a) and ZnO/MS nanocomposite (b).

Fig 1. shows the HR-TEM pictures of MS and ZnO/MS nanocomposite. The embedding of ZnO

SOLID STATE PHYSICS: Proceedings of the 57th DAE Solid State Physics Symposium 2012
AIP Conf. Proc. 1512, 204-205 (2013); doi: 10.1063/1.4790982

nanoparticles in MS is evident from these pictures. Only few particles that are grown to large sizes are appearing as black dots in the fig 1(b). Though the existence of ZnO in the composite is clear but the exact particle size cannot be estimated from this picture. It is inferred from fig 1 (b) that almost all the particles are smaller than 3 nm. FT-IR spectra of MS and ZnO/MS are shown in fig 1. There are no significant differences between these two spectra except the disappearance of 967 cm^{-1} band in nanocomposite. This result indicates that there is no structural collapse of MS with the incorporation of ZnO.

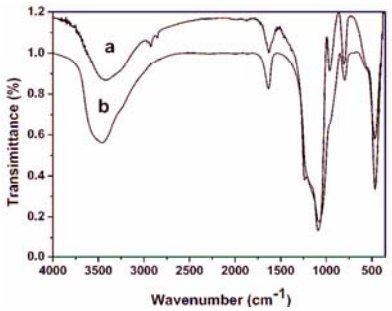

FIGURE 2. Infrared spectra of MS (a) and ZnO/MS (b).

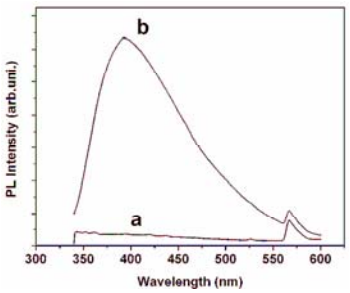

FIGURE 3. PL spectra of MS (a) and ZnO/MS (b).

PL spectra of MS and ZnO/MS nanocomposite are presented in fig 3. MS did not exhibit any luminescence when excited at an excitation wavelength of 320 nm. But the ZnO/MS nanocomposite depicted a strong and broad emission positioned at 393 nm. The explanation for the 568 nm peak was given in reference [5]. With reference to the PL of MS, it can be concluded that emission at 393 nm is solely due to very small ZnO nanoparticles embedded in MS matrix. This result is in good agreement with the earlier reports [5-7]. The generally observed green emission was quenched due to the surface passivation of ZnO nanoparticle. This broad PL band composed of three emissions due to exciton recombination, oxygen vacancies at the ZnO-SiO_2 interface and inner ZnO crystallites [5, 7].

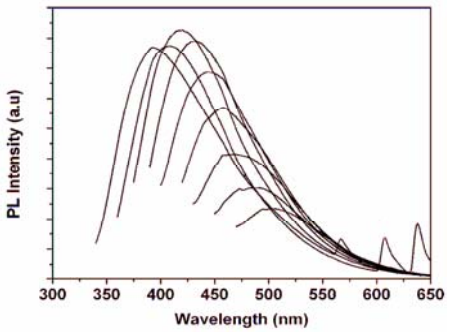

FIGURE 4. PL spectra of ZnO/MS recorded at different excitation wavelengths from 320 nm to 450 nm.

The effect of excitation wavelength on PL of ZnO/MS is shown in fig 4. The position of the PL band is red shifted from 393 nm to 504 nm with increase in the excitation wavelength. But according to Kasha's rule, the same fluorescence emission spectrum is generally observed irrespective of the excitation wavelength. This kind of unusual behavior is termed as red edge effect (REE). The intensity of the emission band does not change appreciably as the excitation wavelength changes from 320 nm to 370 nm. The emission intensity reaches its maximum when excited at 355 nm because it is close to the PL excitation wavelength 347 nm (figure not shown here). But for excitation wavelengths above 370 nm, intensity decreases monotonously. Our previous study confirmed that there is no change in the PL position with increase in particle size [5]. The PLE spectra corresponding to these emission spectra are also red shifted (not shown here). It can infer that quantum confinement effects may be one of the reasons for this REE. Further study is required to obtain insight into this aspect.

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